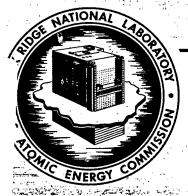
ORNL-4451 DCV UC-70 - Waste Disposal and Processing

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SITING OF FUEL REPROCESSING PLANTS AND
WASTE MANAGEMENT FACILITIES



OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

U.S. ATOMIC ENERGY COMMISSION

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8. HEALTH AND SAFETY ASPECTS OF PLANT SITING

The principal criterion for judging the adequacy of a site for a fuel reprocessing plant is the provision that no undue risk exists with regard to public health and safety in the surrounding areas. Present and fore-seeable technology requires that such plants routinely discharge small quantities of radioactive materials to the atmosphere; for this reason, and also because of the large inventory of physiologically hazardous materials, there is always a small, but finite, probability of a more massive discharge. The magnitude of the routine discharge and the probability of a more massive discharge are determined by the inventory of radioactive materials and by the design features of the plant.

Present licensing procedures for fuel reprocessing plants apply existing federal regulations for radiation protection (10CFR20), licensing of production and utilization facilities (10CFR50),2 and siting of nuclear reactors (10CFR100), 3 wherever applicable, to the plant under study. The safety of a proposed facility is determined by evaluating, as a unit, the proposed plant and the site. The design features of the plant, together with the geological, hydrological, seismological, and meteorological characteristics of the site, are analyzed to determine whether the proposed design is adequate to maintain the barrier between radioactivity and the surrounding population under adverse environmental conditions such as earthquakes, tornados, and floods. The consequences of releasing radioactive effluents during normal operations as well as during "upper limit accident" conditions, are evaluated using environmental characteristics of the site. The calculated concentrations of normal plant effluents are compared with the values published in lOCFR20; the engineered features for prevention and mitigation of the consequences of accidents are compared with the guidelines of lOCFR50; and the calculated doses received by a member of the general public from postulated accidental releases are compared with the guidelines specified in lOCFR100. If, by employing conservative assumptions, it can be demonstrated that engineered safety features and releases under all credible conditions are within the guidelines, then the plant and the site are considered acceptable The following sections of this chapter present estimates of the effect of health and safety considerations on the siting of spent-fuel processing plants. These include the consequences of an expanding nuclear economy on the worldwide distribution of long-lived volatile radionuclides, local environmental effects of the routine release of radionuclides, and the effects of credible accidents.

Section 8.1 presents estimates of the worldwide distribution of ⁸⁵Kr and ³H in an expanding nuclear economy, assuming that these nuclides are released quantitatively to the atmosphere and the hydrosphere. These estimates, together with those of following sections, lead to the conclusion that worldwide pollution hazards will be avoided and local operating personnel will be protected by the necessary expedient of providing engineered safety features and site boundary distances that ensure appropriately low radiation exposures of members of the public at the site boundary.

Section 8.2 presents estimates of the effect of routine releases of radioactive materials from spent-fuel processing plants. The consequences of, and site boundary distances dictated by, routine releases from fuel processing plants were estimated assuming (1) ORML meteorological conditions, (2) the complete release of noble gases and tritium, (3) iodine decontamination factors of 2000 (present technology) and 10⁷ in plants for processing highly irradiated fuels that have decayed 150 and 30 days, respectively, and (4) a particulate-release-rate model that agrees satisfactorily with existing data. For reference purposes, the acceptable concentrations at the site boundary were selected as one-third of the air concentrations listed in 10CFR20, Appendix B, Table II, Column 1, with the exception that the ¹³¹I concentrations were further reduced by a factor of 700 to account for the grass-cow-milk pathway to the thyroids of small children.

The downwind consequences resulting from the routine release of radionuclides from a plant processing light-water reactor (LWR) fuel (postirradiation decay period of 150 days) or a plant processing fast breeder reactor (FBR) fuel (decay time of 30 days) are estimated to be controlled by the release of noble gases and iodine. It is concluded that equipment for removing 50 to 99% of the noble gases is necessary in plants of capacity more than a few tons per day; more efficient iodine removal than that demonstrated in present technology is required for LWR plants of capacity greater than about 6 to 10 tons/day, whereas DF's for iodine as high as 10^8 may be required for FBR plants.

Section 8.3 presents estimates of the effect of releases of radioactive effluents in "upper limit accidents." The consequences of upper
limit accidents were estimated assuming that the acceptable annual dose
commitments resulting from exposure to the cloud or inhalation at the site
boundary are those recommended by the National Committee on Radiation Protection for annual occupational exposure. Although the assumed acceptable
dose commitments have been employed only for reference purposes, they may
be plausible on the basis that the ratio of benefit to probability of
exposure is believed to be greater for an individual of the general population living near the site boundary than for a worker in the plant.

The meteorological and dose commitment analysis was based on the assumptions of flat downwind terrain and exposure to the radioactive cloud. The consequences of downwind ground contamination and additional exposures by such phenomena as reentrainment were not considered as mechanisms that would limit plant siting. Excessive levels of ground contamination would cause inconveniences, require expensive decontamination procedures, and result in property loss; however, they would probably not present an unavoidable threat to the health and safety of the public.

It is concluded that the confinement and ventilation systems in spent-fuel processing plants remove particulates of nonvolatiles dispersed under accidental conditions to such an extent that the upper limit accidents are controlled by the release of such volatile and semivolatile materials as the noble gases, iodine, ruthenium, cesium, and tellurium. Credible upper limit accidents in well-designed facilities for interim storage of wastes, either in liquid or solid form, are estimated to be inconsequential with respect to those from processing operations in the plant.

8.1 Buildup of 85Kr and 3H in an Expanding Nuclear Power Industry

As the free world's nuclear power production increases, the buildup of ⁸⁵Kr in the atmosphere and ³H in the hydrosphere may become important. Therefore, estimates of dose equivalents to the year 2000 from a uniform worldwide distribution of these radionuclides have been made.

Estimates of the annual production of ⁸⁵Kr and ³H are based on the AEC's projected civilian nuclear power economy in the United States and in the free world. ^{4,5} In Fig. 8.1, which shows the growth of the nuclear power industry, foreign capacity in the year 2000 is assumed to be equal to the estimates of capacity in the United States at that time. Thermal power generation was estimated by assuming load factors of 0.8 to 1980 and 0.7 at the year 2000, and a thermal efficiency of 0.31. Thus, in the year 2000, the free world's nuclear capacity for continuous operation is estimated to be 1 million electrical megawatts and 3.3 million thermal megawatts.

The rates of production and accumulation of ⁸⁵Kr and ³H are shown in Fig. 8.2. Production rates were based on an assumed core irradiation of 20,000 Mwd/metric ton and a specific power of 25 Mw/metric ton. The accumulated quantities of ⁸⁵Kr and ³H were obtained by allowing each radionuclide produced in the immediately preceding 5-year period to decay for 2.5 years and adding this value to the previously accumulated quantity (corrected for decay for 5 years). Accordingly, in the year 2000, ⁸⁵Kr production will be 520 megacuries/year, and 3000 megacuries will have been accumulated. Tritium production will be 15 megacuries/year, and 96 megacuries will have been accumulated.

8.1.1 85 Kr Distribution and Dose Equivalent

The concentration of ⁸⁵Kr in the atmosphere was estimated by assuming complete mixing of the ⁸⁵Kr and the air throughout the first 8 miles of the atmosphere. Within this zone, ⁸⁵Kr was assumed to be distributed according to the density mass of air. Above 8 miles, the tropopause would inhibit rapid mixing into the stratosphere. ⁶Rainout was considered negligible, since calculations indicated that the atmosphere

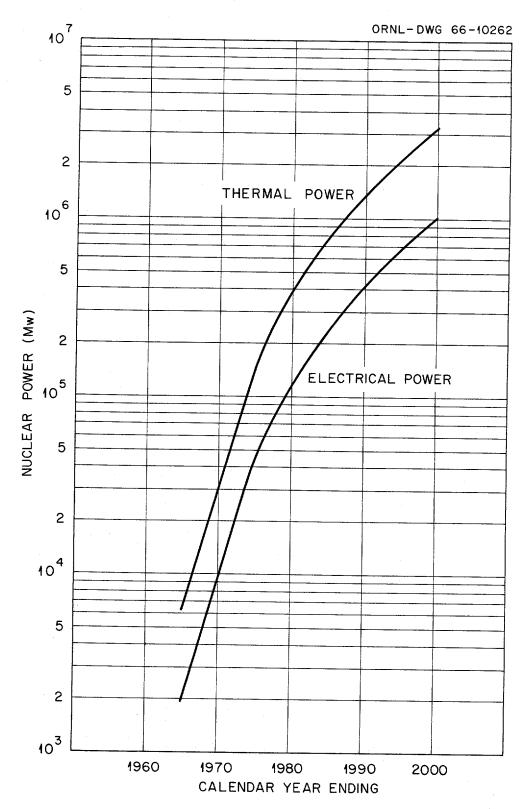


Fig. 8.1. Estimated Growth of Civilian Nuclear Power in the Free World.

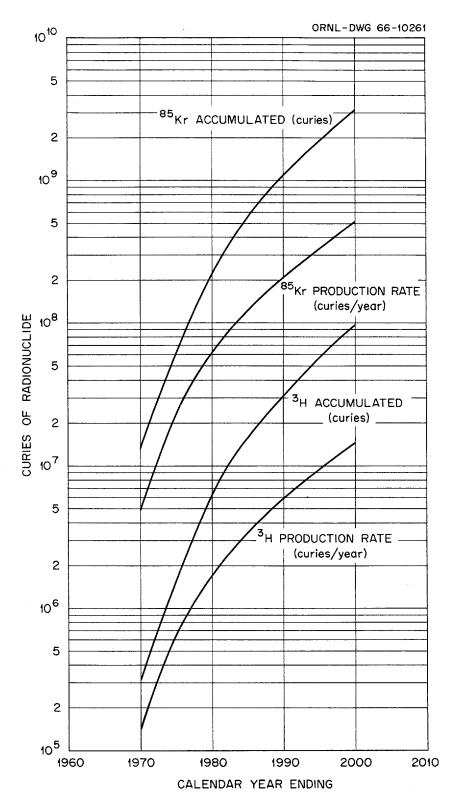


Fig. 8.2. Estimated Production of $^{85}\mathrm{Kr}$ and $^{3}\mathrm{H}$ from the Nuclear Power Industry of the Free World.

contained more than 95% of the stable krypton as compared with the oceans. 7

Figure 8.3 shows the estimated whole-body exposure from 85 Kr as a function of elevation. A maximum dose rate of 1.8 millirems/year in the first one-fourth mile of the atmosphere can be compared to an average background radiation of 100 millirads/year (to skin) near sea level and to permissible whole-body exposures of average population groups of 170 millirems/year, and of members of the public of 500 millirems/year, as recommended by ICRP and FRC. 8,9

8.1.2 Tritium Distribution and Dose Equivalents

Practically all of the tritium in irradiated fuel elements may be released to the environment during spent fuel processing. This release is assumed to occur as HTO, either as tritiated water or as tritiated water vapor. The volumes of circulating waters in the world, listed in Table 8.1, were used to calculate the concentration of tritium in the environment. It was assumed that: (1) tritium was mixed in oceans and seas to a depth of 40 m, (2) all the water in stream channels and in the first 10 km of the atmosphere was circulating, (3) only the portion of the groundwater located in the root zone was available for mixing, and (4) complete isotopic dilution occurred in these waters. As shown in Fig. 8.4, the estimated dose equivalents to body tissue due to inhalation of air and absorption through skin, and to ingestion of surface water containing ${}^{3}\text{H}$ are 7.2 x 10^{-14} and 1.4 x 10^{-3} millirems/year, respectively, for the year 2000. Nonuniform distribution of ³H in rainwater and surface water has been indicated by Libby in his claim that 50% of the tritium released from the detonation of thermonuclear devices in 1958 had fallen between 30° and 50° north latitude. 10 If this occurs in the case of releases of ³H from fuel reprocessing plants, then approximately 10% of the earth's surface will receive one-half of the total 3H. Thus, the dose equivalents in this temperate zone may be five times the calculated average.

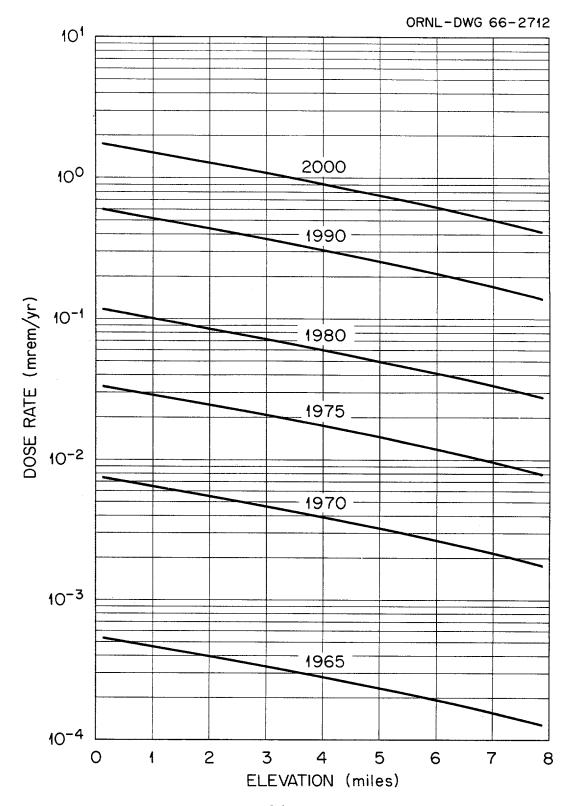


Fig. 8.3. Dose Rate from $^{85}\mathrm{Kr}$ in the Atmosphere According to Elevation.

Table 8.1. Volumes of Circulating Water in the World

	Volume of Water (m ³)	
	Total	North Latitude (30°-50°)
Oceans and seas, in surface 40 m	1.44 x 10 ¹⁶	1.43 x 10 ¹⁵
Stream channels, average	1.17 x 10 ¹³	2.51 x 10 ¹²
Atmospheric moisture, average	1.29 x 10 ¹¹	1.72 x 10 ¹³
Subsurface water in the root zone	2.50 x 10 ¹⁴	5.38 x 10 ¹³
Total circulating water	1.48 x 10 ¹⁶	1.50 x 10 ¹⁵
Total circulating water	1.48 x 10 ⁻³	1.50 x 10 ⁻⁷

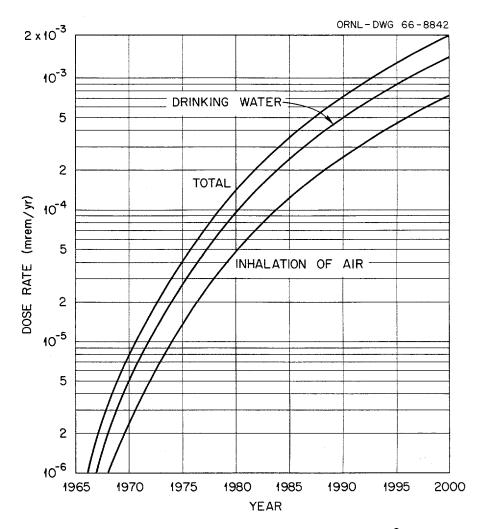


Fig. 8.4. Dose Rate Received by Body Tissue from $^3\mathrm{H}$ That Is Inhaled and Ingested in Drinking Water.

8.2 Routine Release of Radionuclides to the Atmosphere

Present technology requires that fuel reprocessing plants continuously discharge off-gas and ventilation air to the atmosphere. Nonradioactive gases are generated in some process operations; for example, air is supplied deliberately to some process vessels for such purposes as pneumatic liquid level determination, mixing of solutions by sparging, and maintaining nonflammable concentrations of gases and vapors. Since absolutely leak-tight containment barriers are impractical, a flow of ventilation air from normal working areas to enclosures (glove boxes, cells, canyons, etc.) containing radioactive materials in process equipment is required to maintain a contamination gradient. By a variety of mechanisms, radioactive gases, vapors, and aerosols of liquid and solid particles tend to become entrained in these off-gas and ventilation streams. The absolute removal of all radioactive materials from these streams prior to discharge to the atmosphere is impractical.

The policy for the routine discharge of radioactive effluents to the environment is to maintain the rate of release of radioactive materials at the lowest practical level consistent with current technology by careful control and continuous monitoring. In any event, the consequences of the release must be within the limits established by federal regulations (10CFR20), which have the intent of providing that negligible risk to the health and safety of the public will result. This policy is achieved by (1) striving to maintain process vessel enclosures free of mobile radioactive materials in order to minimize the possibility that the ventilation air will become contaminated, (2) maintaining the flow rate of the off-gas that contains (or comes in contact with) mobile radioactive materials at the minimum practical level, (3) employing devices such as scrubbers and filters to remove as much of the radioactive material from the effluent as is practical, and (4) discharging the effluent through stacks to provide effective atmospheric dispersal.

8.2.1 Sources of Routine Releases

The rate of routine release of radionuclides to the atmosphere from fuel reprocessing plants as a function of capacity (Table 8.2) was esti-

Table 8.2. Estimated Routine Release Rates for Radionuclides as a Function of Reprocessing Plant Capacity

	Release Rate per	Unit of Throughput
	LWR Fuel Reprocessing Plant ^a	FBR Fuel Reprocessing Plant ^b
Noble Gas ⁸⁵ Kr ¹³³ Xe	1.0	1.0 0.1
Tritium	1.0	1.0
Halogens	0.001	10-7
Particulates c	1.2 x 10 ⁻⁸	8.5×10^{-10}

^aLWR fuel irradiated to a burnup of 33,000 Mwd/metric ton, at a specific power of 30 Mw/metric ton, and allowed to decay for 150 days. Off-gas rate = 1000 cfm per metric ton per day. Filter effluent = 0.0012 mg of solution per m³. Solution concentration = 0.3 kg of fuel per liter.

bLMFBR (mixed core and blankets) irradiated to a burnup of 33,000 Mwd/metric ton, at a specific power of 58.2 Mw/metric ton, and allowed to decay for 30 days. Off-gas rate = 70 cfm per metric ton per day. Filter effluent = 0.0012 mg of solution per m³. Solution concentration = 0.3 kg of fuel per liter.

CParticulate release rates are assumed to scale approximately as the 0.6 power of the plant throughput rate. The rates given are estimated for a plant with a capacity of 260 metric tons per year.

mated, based on current technology for LWR fuel reprocessing plants and foreseeable technological developments for plants that will process FBR fuels. The corresponding release rates, in curies, may be obtained as the product of the fractional release (Table 8.2), the fuel processing rate (in metric tons/day), and the concentration of the isotopes in a metric ton of fuel (Table 8.3). These values permit a preliminary estimation of site sizes that would result from the effect of routine releases. Section 8.4 will present an analysis of tradeoffs that can be made in site size through the use of additional engineered safety features.

Noble Gases. - A total of approximately 0.001 ft³ (STP) of the noble gases He, Kr, and Xe is generated in each megawatt-day of reactor operation. The radioisotopes of physiological hazard significance that remain after 30 or more days of postirradiation decay are ⁸⁵Kr and ¹³³Xe. Unvented fuel contains approximately 0.3 curie of ⁸⁵Kr for each megawatt-day of burnup. Unvented fuel contains about 1300 curies of ¹³³Xe per megawatt of thermal power after 30 days of decay and negligible quantities after 60 days of decay.

In preparing Table 8.2, it was assumed that these gases will continue to be released quantitatively from LWR fuel reprocessing plants as the fuel is chopped and/or dissolved. It was assumed that, in plants for reprocessing FBR fuels after 30 days of decay, the gas would be held up (in a charcoal bed) for a period of 18 days to effect an order-of-magnitude reduction in the \$133\$Xe activity. Several processes (employing charcoal adsorption, liquid nitrogen, Amsco, or fluorocarbon scrubbing, or permselective membranes), within moderate extensions of current technology, may be employed to remove 90 to 99% of both xenon and krypton if required because of particular site limitations or a strict adherence to a policy of maintaining "lowest practicable" release rates. Release rates, particularly for \$5\$Kr, would be lower for reactor fuels that use the vented fuel concept.

Tritium. - Approximately 0.025 curie of ³H is formed for each megawatt-day of reactor exposure. The common and most stable compound, HTO, is practically unrecoverable by present technology after it has been mixed with water. Present plants discharge tritium essentially quanti-

Table 8.3. Radionuclide Content of LWR Fuel Decayed 150 Days and Mixed Core-Blanket LMFBR Fuel Decayed 30 Days

Concentration (curies/metric ton)				Concentration (curies/metric ton)	
Nuclide	In LWR Fuel	In LMFBR Fuel	Nuclide	In LWR Fuel	In LMFBR Fuel
3 _H	692	932	131 _I	2.17	139,000
85 _{Kr}	11,200	10,200	132 _I	-	4300
89 _{Sr}	96,000	637,000	133 _{Xe}	-	74,400
⁹⁰ Sr	76,600	43,400	134 _{Cs}	213,000	29,000
90 _Y	76,600	43,500	136 _{Cs}	20.8	28,800
91 _Y	159,000	921,000	137 _{Cs}	106,000	109,000
95 _{Zr}	276,000	2,100,000	140 _{Ba}	430	523,000
95 _{Nb}	518,000	2,660,000	140 _{T.a}	495	601,000
99 _{Mo}	-	1810	141 _{Ce}	56,700	1,480,000
99m _{Tc}	-	1730	1կկ _{Ce}	770,000	1,280,000
99 _{Tc}	14.2	14.9	$143_{ m Pr}$	694	6ЦЦ, 0 00
103 _{Ru}	89,100	1,760,000	147 _{Nd}	51.0	185,000
106 _{Ru}	410,000	1,290,000	147_{Pm}	99,400	353,000
103m _{Rh}	89,100	1,760,000	149 _{Pm}	-	61.5
111 _{Ag}	-	12,600	151 _{Sm}	1150	4690
115m _{Cd}	44.3	269	$^{152}\mathrm{Eu}$	11.5	10.5
124 _{Sb}	86.3	76.7	155 $_{ m Eu}$	6370	79,400
125 _{Sn}	20.0	6720	160 _{Tb}	300	9460
125 _{Sb}	8130	19,600	239 _{Np}	17.4	7220
125m _{Te}	3280	6860	238 _{Pu}	2810	11,200
127m Te	6180	61,100	239 _{Pu}	330	3530
127 _{Te}	6110	61,800	240 _{Pu}	478	4260
129m _{Te}	6690	181,000	241 _{Pu}	115,000	600,000
129 _{Te}	4290	116,000	241 _{Am}	200	1570
132 _{Te}	_	41.70	242 _{Cm}	15,000	65,500
129 _I	0.038	0.053	5777 Cm	2490	1240

^aThese data are taken from Tables 3.9, 3.15, 3.33, and 3.39.

tatively to the environment in off-gas and low-level liquid waste. 11 Complete release of tritium to the atmosphere, the planned means of disposal at the MFRP plant, 12 is assumed in Table 8.2. Advanced technology, employing either vented fuel elements or a high-temperature oxidation process after the fuel has been chopped, may reduce the rate of release of tritium from fuel processing plants by factors of 10 to 100.

Halogens. - Of the fission-product halogens, only the isotopes ¹³¹I and ¹²⁹I are physiologically significant after 30 days or more of post-irradiation decay. The ¹³¹I contents of reactor fuels are approximately 0.07 and 2400 curies per megawatt of thermal power after decay times of 150 and 30 days respectively. The ¹²⁹I content is about 10⁻⁶ per megawatt-day of fuel exposure.

In current technology, iodine reports, almost completely, to off-gas systems as I₂, HI, or iodine-organic compounds that are generated in such process operations as chopping, dissolving, and evaporation. Current off-gas trains use caustic scrubbers, which remove approximately 90% of the iodine, and silver nitrate towers, which remove about 99% of the remaining iodine. Through 1962, such devices were used to maintain an average ¹³¹I release rate to the atmosphere of approximately 0.3 curie/day at NRTS, HAPO, SRP, and ORNL. ¹³

It is assumed that plants for reprocessing fuels that have decayed at least 150 days will routinely release 0.1% of the iodine. However, plants for reprocessing fuels after a decay period of 30 days will require development of techniques for maintaining the fractional ¹³¹I release in the range of 10⁻⁷.

Particulates. - The common chemical forms of the fission products other than the noble gases, tritium, and halogens have sufficiently low vapor pressures that the predominant mechanism of release to the off-gas systems is by entrainment of particulates. While several semivolatile fission products (Tc, Se, Ru, Cs, and Te) are known to concentrate in off-gases from certain process operations, the general experience at ORNL in fuel reprocessing operations has been that particulates in off-gas streams have essentially the same relative content of fission products as

the fuel being processed. The explanation is that most of the aerosol in the ventilation streams consists of liquid particles that have become entrained in off-gases that have contacted radioactive solutions. The liquid particles probably have the same fission product content as the original solution since the off-gas streams generally have high relative humidities. The particles that dry after being deposited on ventilation ducts and filters largely tend to remain fixed and to contribute little to the routine release of nonvolatile fission products. (However, they may be the source of a serious accidental release if there is a means for sudden and massive reentrainment.)

At ORNL it has been found that the off-gases from aqueous fuel reprocessing operations contain particles of aqueous solutions at a concentration of approximately 10 mg/m^3 (i.e., the concentration of water particles in fog) and that there are equal weight fractions of particles in the size ranges less than 0.4 μ , 0.4 to 1.3 μ , 3 to 5 μ , and greater than 5 μ . 15 Also, it is known that the weight distribution of particles less than about $5 \,\mu$ in size is relatively constant even if there is gross entrainment of larger particles. Typical deep-bed sand or High Efficiency Particulate Air (HEPA) filters used in processing plants would quantitatively remove 100% of the particles greater than about 3 u in size and about 99.98% of the particles less than 3 u, which have the size distribution indicated above. From these data, it is estimated that the concentration of aerosol in the filter effluent is of the order of 0.0012 mg/m³. Assuming that the radioactive solutions in the plant contain 300 g of fuel per liter (typical of the dissolver and accountability tanks, which contribute significantly to the off-gas) and have a specific gravity of about 1.2, the estimated concentration of fuel in the filter effluent is 0.3×10^{-12} metric ton of fuel per cubic meter of air.

The estimated fractional release of fuel to the atmosphere from a 1-metric ton/day plant for processing \geq 150-day-decayed LWR fuel, using current technology, is 1.2 x 10⁻⁸, assuming a combined dissolver and vessel off-gas flow rate of 1000 cfm. By comparison, the dissolver and the vessel off-gas flow rates are 400 and 620 cfm, respectively, at the NFS plant 16 and approximately 500 and 1000 cfm at the Hanford Purex plant.

It is estimated that the flow rate of the dissolver-vessel off-gas at the MFRP plant will be 250 cfm. 12

The estimated fractional release from a 1-metric ton/day plant corresponds to daily release rates of 0.037 curie of mixed fission products, 0.0006 curie of 90 Sr, 0.007 curie of 95 Zr- 95 Nb, 0.004 curie of 106 Ru, 0.0005 curie of 11 Ce, and 0.00003 curie of Pu. By comparison, the average daily release of nonvolatile fission products from the three Hanford processing plant stacks includes 0.01 curie of 95 Zr- 95 Nb, 0.007 curie of 103 Ru, 0.006 curie of 106 Ru, 0.001 curie of 111 Ce, and 0.00003 curie of total alpha emitters (presumed to be Pu). The is estimated that the daily release of particulates from the MFRP plant stack will consist of less than 0.006 curie of mixed fission products and less than 0.002 curie of alpha activity from plutonium. The estimated daily release of particulates from the 5-metric ton/day BNFP plant consists of less than 0.17 curie of mixed fission products and less than 0.0001 curie of alpha activity from plutonium; this corresponds to a fractional release of about 1 x 10 8.

It is estimated that technological developments will permit the dissolver and the vessel off-gas flow rates to be reduced to 20 and 50 cfm in 1-metric ton/day plants that would process 30-day-decayed FBR fuel. If such is the case, the routine release of particulate activity should be lower than from current plants, in spite of the higher specific activity of FBR fuels.

It is assumed that the routine release of radioactive particulates to the environment will increase in direct proportion to the vessel off-gas flow rate in plants having larger throughput rates. The fuel inventory of individual process vessels will not increase in direct proportion to the production rate because of the necessity for multiple equipment lines to permit continuity of operation and the use of progressively more continuous equipment. The routine release to the off-gas system is roughly proportional to the area of the interface between the radioactive solid or solution and the gas. Radioactive aerosols are entrained in off-gas streams primarily by sparging (usually at a fixed rate of approximately l scfm/ft²), but also by diffusion and recoil from surfaces. As a first

approximation, continuous equipment will have a greater surface-to-volume ratio, which will offset the effect of larger process vessels.

8.2.2 Local Environmental Consequences of Releasing 85 Kr and 3 H

Many pathways have been postulated by which radionuclides may be transmitted through the environment and thereby contribute to the total dose received by man. 19 A generalized model that relates the principal parameters involved in estimating the external dose is as follows: 20

$$D_{ijk}^{ext} [t_1, t_2, \gamma(t_1)] = Q_{ij} \int_{0}^{t_2} P_{ijk} (t) C_{ij} [\gamma(t)] dt, \qquad (1)$$

where

 D_{ijk}^{ext} [t₁, t₂, γ (t₁)] = total external dose to radionuclide i in pathway j at location k for an individual of age γ (t₁) at the beginning of exposure,

P_{ijk}(t) = concentration of radionuclide i in pathway j at location k during time t per unit of radionuclide initially available, and

 $C_{ij}[\gamma(t)]$ = dose rate to the reference organ of an individual of age γ per unit concentration of radionuclide.

The total external dose due to radionuclide i in pathway j at location k, accumulated from time t_1 to t_2 by an individual of age $\gamma(t_1)$ at the beginning of exposure, is the integral of the product of the level of contamination (the quantity Q_{ij} and the concentration P_{ijk}) and the dose rate term, C_{ij} . The later term includes all necessary factors that account for the habits and characteristics of the individual. With minor changes, the same expression can be used to estimate internal dose. For internal dose, the C_{ij} term denotes the dose commitment in the (t_2-t) days following a one-day exposure of the individual.

According to the International Commission on Radiological Protection, the entire human body is the critical organ for exposure to $^{85}\mathrm{Kr.}^8$ The principal mode of exposure is submersion in contaminated air. Body tissue is the critical organ in the case of exposure to tritium as tritiated water or tritiated water vapor. However, the external dose resulting from submersion in air containing HTO vapor is limited to areas where the skin has minimal thickness, because of the limited penetration range of tritium's beta particle.

Prior studies at Hanford and Oak Ridge have demonstrated that not all modes of exposure, or pathways contributing to the same mode, are of equal importance. The modes of exposure considered in this analysis will include ingestion, inhalation (and accompanying skin absorption), irradiation from a contaminated surface, submersion in contaminated water, and submersion in contaminated air. These estimates of dose consider only the dose to "standard" man.

Procedures for Estimating Permissible Release. - Acceptable release rates for ⁸⁵Kr and ³H were investigated for a hypothetical fuel reprocessing plant located at the Oak Ridge National Laboratory. This selection was made since information was already available on some of the environmental factors that influence the dispersion and possible reconcentration of fission products that may be released.

Average annual downwind air concentrations are calculated by a modified Gaussian plume formula as follows:

$$X(9x) = \sum_{i=1}^{S} \frac{R}{\sigma(Sx)_{z}} \frac{2.032F(9S)_{i}Q_{o}}{\sigma(Sx)_{z} \overline{\mu}(9S)_{i}x} \cdot \exp\left[-\frac{h^{2}}{2\sigma(Sx)_{z}^{2}}\right], \qquad (2)$$

where

 $X(\theta x)$ = average annual concentration along a 22.5° arc at distance x in direction 9 (curies/m³),

 $F(\theta S)_i$ = fraction of time that the wind is in direction θ , for stability S and wind speed group i,

Q = initial emission rate (curies/sec),

 $\sigma(Sx)_{Z}$ = vertical dispersion coefficient at distance x for stability S (m),

 $\bar{u}(\theta S)_{\hat{1}}$ = average wind speed in direction θ , for stability S and speed group i (m/sec),

h = stack height (m),

R = index denoting wind-speed groups,

S = index denoting stability parameter.

This expression is obtained by integrating the Gaussian plume formula over the crosswind direction and distributing the results uniformly along the entire arc. Since the average wind-speed vector and its frequency of occurrence are used, calculations yield average annual air concentrations. Applications of this technique have been demonstrated previously by Culkowski. ²³

Equation (2) is modified to include washout and fallout by multiplying by the appropriate correction factors. Corrections for washout and fallout are based on the work of Chamberlain and Slade respectively. These corrections are as follows:

$$Q_{\text{washout}} = \exp \left[-\frac{\Lambda^{X}}{\mu(\theta S)_{i}} \right] , \qquad (3)$$

where Λ is the washout coefficient (sec⁻¹); and

$$Q_{\text{fallout}} = \exp \left\{-\left(\frac{2}{\pi}\right)^{1/2} \frac{V_{\text{g}}}{\overline{\mu}(\theta S)_{i}} \int_{0}^{x} \frac{1}{\sigma(Sx)_{z}} \exp \left[-\frac{h^{2}}{2\sigma(Sx)^{2}}\right] dx\right\}, (4)$$

where V_g is the deposition velocity (m/sec). Equation (4) can be evaluated numerically, based on curves of σ_z values given by Hilsmeier and Gifford. ²⁶

Figure 8.5 shows the calculated air concentrations at the ground surface for a 1-part/sec release from a 100-m stack located at ORNL. The most recent meteorological data reported by Hilsmeier are used in these calculations. ²⁷ Concentrations shown in Fig. 8.5 can be compared with others that include fallout, washout, and changes in stack height; by this process, average annual doses can be estimated for a variety of conditions.

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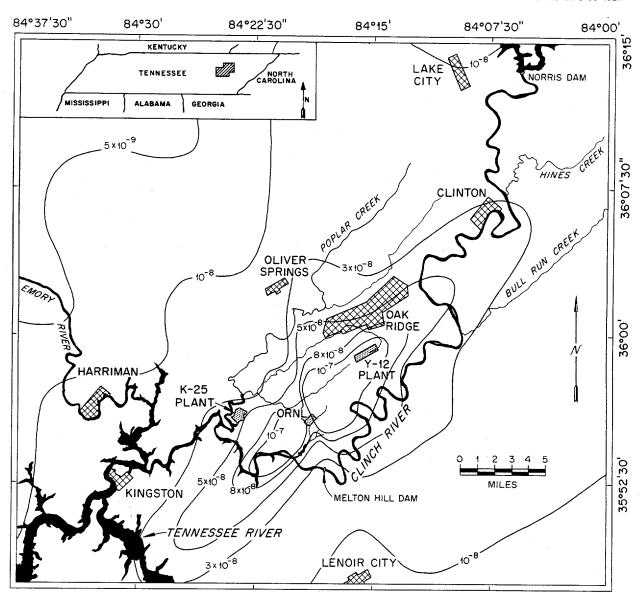


Fig. 8.5. Average Annual Air Concentrations at Ground Surface in Parts per Cubic Meter. Source height, h = 100 m; source strength, Q = 1 part/sec.

Washout. - Washout coefficients for soluble gases have been calculated by Chamberlain, using the assumption that the rate of absorption is controlled by the rate of gas diffusion to the raindrop. 24,28 Since the solubility of krypton in water is small (1.85 x 10⁻¹⁰ g of krypton per gram of water at equilibrium), 7 it was assumed that the solubility limit controls the amount of krypton absorbed. The solubility of 85Kr in rainwater, even when released at 1 curie/sec, would be limited by the stable krypton in the atmosphere (about 4×10^{-3} g/m³ near sea level).²⁹ further assumed that krypton is washed out of the atmosphere, beginning at an average height of 1 mile. This assumption is based on the height of rain-bearing cumulus clouds and on the extent of vertical development of radioactive clouds released as a point source. The average intensity of rainfall is about 4 mm/hr in the Oak Ridge area, 30 and, at equilibrium, 2 x 10 -14 g of krypton per second could be absorbed in a column of the atmosphere 1 mile high and 1 cm2 in area. About 5 x 10-4 g of stable krypton per square centimeter is contained in the atmosphere to a height of 1 mile. Based on these considerations, the average washout coefficient was calculated to be:

$$\Lambda = \frac{2 \times 10^{-14} \text{ g of Kr sec}^{-1} \text{ cm}^{-2}}{5 \times 10^{-14} \text{ g of Kr cm}^{-2}} = 4 \times 10^{-11} \text{ sec}^{-1}.$$

The washout coefficient of tritiated water vapor (HTO) has been estimated from Chamberlain's calculations for SO_2 deposition in rainwater. It was considered to be proportional to the diffusion coefficient of the vapor in air. Therefore, the following expression was used to calculate $\Lambda_{\rm HTO}$ for a 4-mm/hr rainfall:

$$\Lambda_{\rm HTO} = \Lambda_{\rm SO_2} \frac{D_{\rm HTO}}{D_{\rm SO_2}} = 4 \times 10^{-4} \, {\rm sec}^{-1}$$
,

where

$$\begin{split} &\Lambda_{\rm H\,IO} = {\rm washout\ coefficient\ of\ HTO\ vapor\ (sec^{-1}),} \\ &\Lambda_{\rm SO_2} = {\rm washout\ coefficient\ of\ SO_2\ (2\ x\ 10^{-4}\ sec^{-1}),}^{24} \\ &D_{\rm H\,TO} = {\rm diffusion\ coefficient\ of\ HTO\ vapor\ in\ air\ (0.23\ cm^2/sec),}^{31} \\ &D_{\rm SO_2} = {\rm diffusion\ coefficient\ of\ SO_2\ in\ air\ (0.115\ cm^2/sec).}^{24} \end{split}$$

Loss of HTO from a raindrop to the atmosphere was assumed to be negligible. This assumption is valid if the distance the raindrop falls below the contaminated cloud is small as compared with the relaxation length.*

A washout coefficient of 4×10^{-4} sec⁻¹ for HTO vapor is consistent with that indicated by Chamberlain and Eggleton. Similar values can also be deduced from published data on the concentration of tritium in the atmosphere and in rainwater. For example, the maximum concentration of tritium, in tritium units (TU), was reported to be 10^6 in hydrogen, $3^3 \times 10^3$ in water vapor, $3^3 \times 10^4$ in methane, $3^4 \times 10^3$ in rainwater. Assuming the average water content of air to be 8.6 g/m^3 (at 50% relative humidity and 20°C) and using the values of TU listed above, the concentration of tritium in the atmosphere is estimated to be 2.9×10^{-11} curie/m³. The tritium content in a column of the atmosphere 1 mile high and 1 m² in area is 4.7×10^{-8} curie. The rate of tritium removal from a $1-\text{m}^2$ area by a 4-mm/hr rainfall would be 5.1×10^{-12} curie/sec. Therefore, the washout coefficient is calculated to be:

$$\Lambda = \frac{5.1 \times 10^{-12} \text{ curie/sec}}{h.7 \times 10^{-8} \text{ curie}} = 1.1 \times 10^{-h} \text{ sec}^{-1}.$$

Since the annual frequency of a 4-mm/hr rainfall in Oak Ridge is only 0.037, the average annual ground-level air concentrations are not reduced significantly at these washout coefficients.

<u>Fallout.</u> - If the sorption of a radionuclide by the ground surface is irreversible, the flux of the radionuclide to the surface does not depend on the amount already deposited. ³⁶ Chamberlain describes the rate of deposition for such a system in terms of a deposition velocity. The following equation is used to estimate the deposition velocity of gases or very small particles: ²⁸

$$V_g(Z_1) = \frac{ku*}{\ln (ku*Z_1D^{-1})}$$
, (5)

^{*}Relaxation length is the distance in which the isotopic composition of the raindrop decreases by 1/e.

where

 V_g = deposition velocity (cm/sec),

k = von Karman's constant (0.4),

u* = friction velocity (cm/sec),

Z₁ = reference height above ground surface at which the concentration
 of the radionuclide is measured (cm),

D = molecular diffusivity (cm^2/sec) .

By assuming u* = 40 cm/sec (appropriate to the Oak Ridge area), 37 $Z_1 = 100$ cm, and D = 0.15 cm²/sec (diffusion coefficient of krypton in nitrogen), 38 the deposition velocity of krypton is 1.7 cm/sec. For tritiated water vapor, with D = 0.23 cm²/sec, the deposition velocity is 1.8 cm/sec. 30

The retention of krypton by the soil is assumed to be limited by the adsorption capacity of the soil for krypton. The retention of krypton by soil can be estimated, assuming that the amount of adsorbed krypton is proportional to the surface area of the soil. From measurements of krypton adsorption on charcoal (2 x 10^{-6} g of krypton per gram of charcoal at 25°C and 10^{-3} mm Hg partial pressure)³⁹ and the ratio of soil area to charcoal surface area (0.05),40 the adsorption of krypton by soil is estimated to be 10^{-7} g per gram of soil (or 1.2 x 10^{-7} g/cm³ for a soil density of 1.2 g/cm³). The rate at which krypton is deposited on the soil is estimated as the product of the deposition velocity ($V_g = 0.017 \text{ m/sec}$) and the krypton concentration in the atmosphere $(X = 10^{-3} \text{ g/m}^3)$, or $6.8 \times 10^{-5} \text{ g m}^{-2} \text{ sec}^{-1}$. At this rate, the soil will probably become saturated with krypton and may not act as a perfect sink for the addition of $^{85}\mathrm{Kr}$. The amount of $^{85}\mathrm{Kr}$ adsorbed on the soil at equilibrium is assumed to be directly proportional to the ratio of radioactive and stable krypton in the atmosphere. For a 85Kr release rate of 1 curie/sec, the soil load (at equilibrium) at the point of maximum ground-level air concentration would be the product of 1.2 x 10^{-7} g/cm³ (soil) and 4 x 10^{-9} g/m³ divided by the product of 4×10^{-3} g/m³ (air) and 397 curies per gram of 85Kr, or $4.8 \times 10^{-11} \text{ curie/cm}^3$.

Since the adsorption of 85 Kr by the soil may not be an irreversible process, the net flux of 85 Kr to the soil (g m⁻² sec⁻¹) may change as the

soil approaches saturation. The deposition velocity calculated from Eq. (4) can be used to estimate only the initial flux of 85 Kr to the soil (and cloud depletion by fallout). The flux to the soil would be expected to diminish with time until steady-state conditions are attained.

The mechanisms by which HTO vapor may be retained by the soil would probably include adsorption, condensation, and exchange with soil moisture. Evaporation, evapotranspiration, and soil drainage would act to redistribute the deposited material. Water vapor $(H_{2}0)$ in the atmosphere would also be acted upon by these mechanisms and would compete with HTO for retention by the soil. In the absence of isotopic fractionation, the ratio at equilibrium of the deposition rate of HTO vapor to H2O vapor would be directly proportional to the ratio of their respective concentrations in air. A deposition velocity of 0.018 m/sec can be used to estimate the flux when the soil acts as a perfect sink. Assuming an average water vapor content in the atmosphere of 8.6 g/m³, the flux of water vapor to the soil due to fallout would be 0.15 g m⁻² sec⁻¹ (4.7 x 10^6 g m⁻² year⁻¹). The average rate of rainfall in Oak Ridge is 1.1 g m⁻² sec⁻¹. For a frequency of rainfall of 0.037, the quantity of rainwater deposited each year is $1.3 \times 10^6 \text{ g/m}^2$. These rates imply that, if the soil acts as a perfect sink for water vapor fallout, the soil would receive an amount of water equivalent to a continuous rainfall of about 0.4 mm/hr. Obviously, this does not occur; thus the soil would not act as a perfect sink for either HTO or H20 vapor, and the flux of HTO vapor to the soil would be expected to vary with time. Only a free water surface, such as the Clinch River, can be assumed to act as a perfect sink for HTO vapor that is released from a stack. Further studies are necessary to evaluate the flux of both $^{3}\mathrm{H}$ and $^{85}\mathrm{Kr}$ to the soil during transient and steady-state conditions.

As a first approximation, the following conservative assumptions are made: (1) the contaminated cloud is not depleted of 85 Kr and HTO by fallout; (2) the quantity of 85 Kr retained by the soil or by the Clinch River is proportional to the ratio of radioactive and stable krypton in the atmosphere; (3) the quantity of HTO retained by the soil is proportional to the ratio of HTO vapor and H_2 O vapor in the atmosphere; and (4) the Clinch River is a perfect sink for HTO vapor.

Krypton and $\rm H_2^{\,0}$ vapor may be adsorbed on particles in the atmosphere and, therefore, be deposited on the ground with these particles. The quantity of krypton associated with particles is estimated by assuming that the air contains 1.4×10^{-4} g of particles per cubic meter (average of city atmosphere)⁴¹ and, as an upper limit, that these particles can adsorb as much krypton as charcoal (2 x 10^{-6} g of krypton per gram). Adsorption of krypton on particles is estimated to be 3 x 10^{-10} g per cubic meter of air, which is negligible as compared with the krypton in the atmosphere (4 x 10^{-3} g/m³). Assuming that charcoal particles can retain two layers of water vapor, the adsorption of water vapor by the particles is estimated to be 7 x 10^{-5} g per cubic meter of air. This value is negligible as compared with that of water vapor in the atmosphere (8.6 g/m³).

Dose Estimation Models. - Methods described and parameters given in ICRP Publication 2^8 are used to convert concentrations (X in curies/m³) to estimates of dose equivalents to "standard" man from submersion in a contaminated cloud, from ingestion, and from inhalation. In particular, Eqs. (12), (13), and (20) in ref. 8 are used, and equilibrium conditions are assumed where appropriate. These dose equations are summarized in Table 8.4.

Submersion dose rates in contaminated water were calculated by assuming that the body is in the center of a sphere and receives equal quantities of radiation from all directions. Other assumptions included: (1) the radius of the contaminated fluid is large as compared with the range of beta particles and to the half thickness of the fluid for gamma rays, (2) an effective energy that is equal to the average energy of the beta particle is absorbed, and (3) penetration distance for the beta particle in the body is short, thus limiting beta radiation to skin and subsurface tissue. The following expressions were derived to calculate dose equivalents at the surface of a body submerged in contaminated fluid:

For
$$85_{Kr}$$
:

R = 0.26 X_W rems/hr

For ^{3}H :

R = 1.1 x 10⁻² X_W rems/hr,

Table 8.4. Equations to Calculate Dose Equivalents (rems per week) to Standard Man^a

T-V-C-C-T-T		40-hr Week Exposure	Exposure	168-hr Week Exposure	r Exposure
Mode ^b	Organ	$^{3}\mathrm{H}$	$^{85}\mathrm{Kr}$	$^{3_{ m H}}$	$^{85} m Kr$
Inhalation and skin absorption	Total body	1.2 × 10 ⁴ x _a	1.	3.6 x 10 ⁴ X _a	
Inhalation and skin absorption	Body tissue	2.0 × 10 ⁴ X _a		5.8 × 10 ⁴ x _a	
Ingestion	Total body	0.67 X		1.9 X	
Ingestion	Body tissue	1.1 X _W		3.2 X	
Submersion in air	Total body		$9.2 \times 10^3 \text{ x}_{\text{a}}$		$h.0 \times 10^4 \text{ K}_3$
Submersion in air	Skin	3.9 x 10^2 X _a		$1.7 \times 10^3 \text{ X}_{a}$	3
External exposure, 2.5 ft above con- taminated ground surface	Total body		$2.4 \times 10^{1} \text{ x}_{a}$		$1.0 \times 10^2 \text{ x}_{a}$

a Dose rate, in rems/week, when the concentration in air, x_a , or the concentration in water, x_w , is expressed in units of curies/m³.

b Exposure mode and critical organ for inhalation and skin absorption, ingestion, and submersion in air are based on information contained in ref. 8.

where

 $X_{\rm W}$ is the concentration of $^{85}{\rm Kr}$ or $^{3}{\rm H}$ in the fluid in microcuries per gram of fluid.

Hine and Brownell describe the derivation of equations that relate to the calculation of dose rates in air from beta emitters associated with an infinite plane of negligible thickness. 42 Equations (10), (11), (20), and (21) in ref. 42 are selected for calculation in cases where the energydependent parameters are those adapted for dose estimates in soft tissue. Equation 9-30 from work by Morgan and Turner is used to calculate the dose due to gamma emitters when the source is of infinite planar extent and infinite thickness. 43 External dose equations listed in Table 8.4 for soil contaminated with 85Kr are then derived from the expected soil load $(4.8 \times 10^{-11} \text{ curie/cm}^3)$ at the maximum air concentration $(1.6 \times 10^{-6} \text{ µc/cm}^3)$. The range, in aluminum, of the average-energy beta particle from 85Kr is used to estimate the thickness of contaminated soil contributing to the beta radiation dose and, thus, the amount present per unit area. beta radiation dose rate is calculated by assuming that this amount of 85 Kr is spread uniformly over the surface without taking self-absorption within the soil layer into consideration.

Estimated Dose Equivalents. - For the purposes of this analysis, we have chosen ⁸⁵Kr and ³H release rates of 0.55 and 0.034 curie/sec respectively. These release rates correspond to a reprocessing plant with a capacity of about 6 metric tons/day (a fuel exposure of 33,000 Mwd/metric ton and a specific power of 30 Mw/metric ton). All of the ⁸⁵Kr is assumed to be released to the atmosphere. It is assumed that 0.0085 curie of ³H per second is released to the atmosphere as HTO vapor and 0.0255 curie of ³H per second is discharged to the Clinch River at mile 20.5 (below the Oak Ridge municipal water intake and above the water intake for the Oak Ridge Gaseous Diffusion Plant) as liquid waste and is diluted with 4900 ft³ of river water per second. Other schemes of ³H release, such as the distillation of ³H-bearing liquids and release to the stack as water vapor, are possible, but would require an appropriate adjustment in the dose estimates that follow.

Surface water in the area can be contaminated directly by fallout and washout of 85 Kr and 3 H, as well as by the direct release of HTO in liquid waste. Clinch River water is assumed to equilibrate with 85 Kr at the maximum specific activity expected in the atmosphere up to the solubility limit of krypton in water. Soils that equilibrate with 85 Kr or 3 H from the overlying atmosphere are assumed to retain these materials. The contribution, by washout, is based on the deposition rates that are calculated in the northeast sector; and these are the maximum rates. Assumptions made for the addition of 85 Kr by fallout and 3 H by washout would then give conservative estimates of concentrations in Clinch River water.

Figure 8.6 shows the average annual dose equivalents in millirems per year, to the total body for submersion in air containing ⁸⁵Kr. Exposures are assumed to be continuous (168 hr/week and 50 weeks/year). These average dose rates were calculated from the ground-level air concentrations (Fig. 8.5) that result from a l-curie/sec release rate and a negligible cloud depletion by washout and fallout. Figure 8.7 shows the estimated dose rates for continuous exposure, in millirems per year, at a distance 2.5 ft above a ground surface contaminated with ⁸⁵Kr. Ionizing radiation associated with tritium on the ground surface would be shielded effectively by 2.5 ft of air.

Table 8.5 contains the estimated annual dose equivalents, to the standard man working at the Oak Ridge Gaseous Diffusion Plant (ORGDP) or residing in Oak Ridge, due to the release of 0.55 curie of 85 Kr and 0.034 curie of 3 H per second in the environment. Periods of occupancy are 40 hr/week and 50 weeks/year for the ORGDP employee, and 168 hr/week and 50 weeks/year for the Oak Ridge resident. A "less than" sign preceding certain values reflects a conservative estimate. The critical modes of exposure are submersion in air for 85 Kr, and inhalation and absorption through the skin for 3 H. The estimated total-body exposure, due to releases from a 6-ton/day plant, is about 90 millirems/year for the standard man residing in Oak Ridge.

Interpretation of Results. - The Federal Radiation Council (FRC), in consideration of a linear relationship between biological effect and dose, background radiation, benefits and risks to be derived from radiation use,

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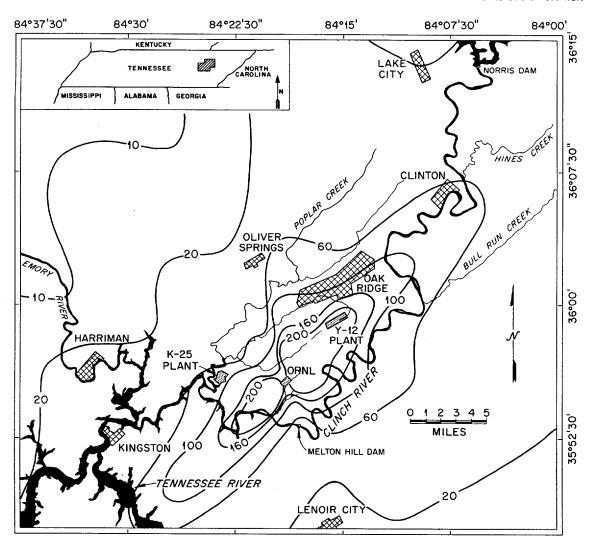


Fig. 8.6. Average Annual Ground-Level Dose Equivalents, in millirems/year, to Total Body for Submersion in Air Containing 85 Kr. Source height, h = 100 m; source strength, Q = 1 curie/sec.

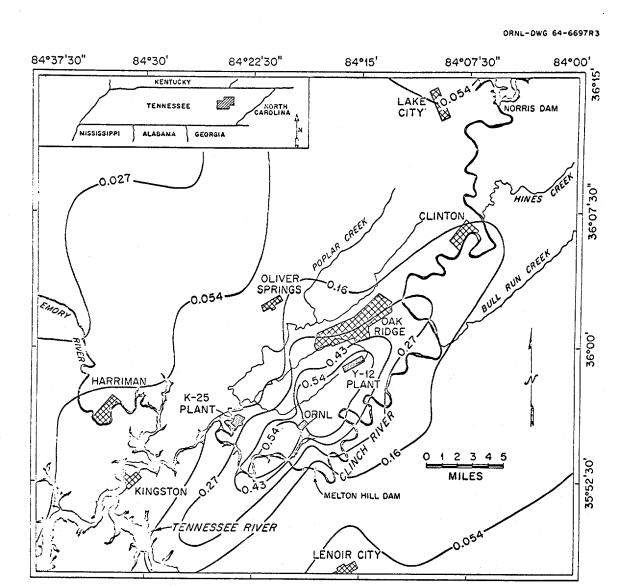


Fig. 8.7. Average Annual Dose Equivalents, in millirems/year, at a Distance 2.5 ft Above Ground Surface Contaminated with 85 Kr. Source height, h = 100 m; source strength, Q = 1 curie/sec.

Table 8.5. Estimated Annual Dose Equivalents, in millirems, Received by the Standard Man due to a 6-metric ton-per-day Reprocessing Plant Located at ORNL

Mode of	Reference Organ	Dose Rate (millirems/year)			
Exposure ^a		Employee of ORGDP	Oak Ridge Resident		
Krypton-85					
Submersion in air	Total body	13	88		
Submersion in water	Total body	<0.006	<0.006		
Contaminated ground (2.5 ft above surface)	Total body	0.03	0.2		
<u>Tritium</u>					
Inhalation and skin absorp-tion	Body tissue	0.43	1.9		
Ingestion of water	Body tissue	10.0	<0.08		
Submersion in air	Skin	0.009	0.06		
Submersion in water	Skin	0.22	<0.001		

^aExposure mode and reference organ for submersion in air, inhalation and skin absorption, and ingestion of water is based on information contained in ref. 8.

and other factors, established, as its basic recommendation, that the annual radiation exposure to the whole bodies of individuals in the general population (exclusive of natural background or medical exposures) should not exceed 0.5 rem. ⁹ In the event of widespread radioactive contamination, and because of uncertainties in the relationship between average and maximum exposure, the FRC suggests the use of the arbitrary assumption that the majority of individuals do not vary from the average by a factor greater than 3. Thus, the use of 0.17 rem for the annual whole-body exposure of average population groups is recommended. When the size of the population group under consideration is sufficiently large, consideration must also be given to the contribution of the genetically significant population dose. According to the FRC,

"The use of 0.17 rem per capita per year, as described in paragraph 5.4 as a technique for assuring that the basic Guide for individual whole body dose is not exceeded, is likely in the immediate future to assure that the gonadal exposure Guide is not exceeded."

These guides are essentially in agreement with current recommendations of the ICRP and NCRP. Each agency also encourages that every reasonable effort be made to keep exposures as far below the offered guidance as practicable.

In current reports, the ICRP and NCRP list the total body as the critical organ and submersion in a semispherical infinite cloud of radioactive gas as the critical mode of exposure for \$85 kr.8,44 However, the basic recommendations in effect at the time these reports were published considered the whole body and the blood-forming organs as a unit, and, as mentioned above, even the genetic dose was partially related to whole-body dose. Because of the rather short range of the beta radiation from \$85 kr, only a small fraction of the total mass of the blood-forming organs or the testes would be exposed to a significant part of the beta dose to skin; however, this might be as much as 1 g of red marrow (e.g., in the skull). The mass of 1 g was previously used as a basis for dose assessment. In later publications of the ICRP, the principle of averaging the dose over organs and tissues is stated without qualification. This principle would permit a higher dose. Since the beta radiation does penetrate well below the skin layer, as shown subsequently, a significant

volume of body tissue would be irradiated at 50% or greater of the surface skin dose. If this tissue is to be limited to 1.5 rems per year, an increase by about a factor of 3 or slightly more might be warranted.

Krypton-85 decays principally by emitting a 0.514-Mev photon 0.7% of the time and a beta particle of 0.695 Mev maximum energy 99.3% of the time. 47 Calculations indicate that the total dose at the surface of a body submerged in a semispherical infinite cloud containing 85Kr is composed of about 99% beta and 1% gamma. The ranges in tissue of the beta rays of maximum and average energy are estimated to be 2.6 mm and 0.55 mm respectively. A considerable fraction of the beta particle energy will be deposited, on the average, in the epidermal (range in thickness, 0.023 to 0.070 mm) and dermal (average thickness, 0.70 mm) layers of the skin of the total body. Thus, there is reason to reevaluate the total body as the critical organ from submersion exposure to 85Kr as a function of depth-dose relationships.

For the complete release of ⁸⁵Kr and ³H from a fuel reprocessing plant sited at ORNL, ⁸⁵Kr would be of greater dose potential to man than ³H. Of the modes of exposure considered, submersion in contaminated air would deliver the largest dose, that is, about 90 millirems per year for a 6-ton/day plant. As explained above, current guidance for total-body exposure to ⁸⁵Kr limits the maximum permissible dose of individuals in the general population to 500 millirems per year (and of average population groups to 170 millirems per year).

The potential dose resulting from the release of ³H in liquid waste is small because credit can be taken for dilution in the Clinch River in which flow is substantial (4919 ft³/sec) and the river is not used as a source of municipal water. Dose estimates by the ingestion of water (10 millirems annually) at ORGDP would increase in direct proportion to a reduction in flow rate and increase by a factor of 3 if the water were used as a municipal water supply. Disposal of ³H in water vapor released to the stack may be one way to reduce the potential exposure from ingestion of water.

Economic benefits would be expected to accrue from large processing plants, but remote siting may not be a practical method for restricting population exposures in the future. This is the justification, therefore, to continue research and development studies, now in progress, to reduce the amounts of 85 Kr and 3 H released and to understand more completely the fate of these radionuclides after discharge to the environment.

8.2.3 Local Environmental Consequences from All Routine Releases

Although the routine releases of ⁸⁵Kr and ³H were emphasized in the preceding section, the absolute removal of all other radioactive materials from gases and vapors prior to discharge to the atmosphere is impractical. Of the remaining radionuclides, ¹³¹I is known to be important because of reconcentration that occurs in the grass-cow-milk pathway to the thyroids of small children. Less experimental information is available on the behavior of ¹²⁹I in the environment, but the assumption will be made that the grass-cow-milk pathway is the dominant mode of exposure from this radioisotope. The controlling pathways for exposure from particulates of mixed fission products and actinides are, also, not well understood. However, it is known that, under some circumstances, such effects as reconcentration in fish or crops and resuspension may be important. In this analysis, it will be assumed that the major exposures from the atmospheric release of particulates will result from direct inhalation of the contaminated air.

Based on the results of the preceding section, it will be assumed that the maximum acceptable average annual concentrations of 85 Kr and 3 H in air at the boundary of a fuel reprocessing plant site are 1 x 10⁻⁷ and 7 x 10⁻⁸ curies/m³ respectively. These are the values recommended by 10CFR2O, Appendix B, Column II, and correspond to annual whole-body exposures of 170 millirems. Experimental evidence has suggested that the average annual concentration of 131 I in air, as provided by 10CFR2O, should be reduced by a factor of about 700 to account for deposition followed by the grass-cow-milk pathway. It is assumed that this same reconcentration factor of 700 should be applied to 129 I, but that, in addition, another factor of 10 is required to account for the relatively longer effective half-life of 129 I on grass. Consequently, the assumed

maximum acceptable average annual concentrations of 131 I and 129 I at the site boundary are 1 x 10 $^{-10}$ /700, or 1.4 x 10 $^{-13}$ curie/m³, and 2 x 10 $^{-11}$ /700/10, or 3 x 10 $^{-15}$ curie/m³, respectively. The assumed acceptable average annual air concentrations of particulates containing mixtures of radionuclides are weighted average values that were derived using one-third of the 10CFR20 concentrations for specific nuclides and relative radionuclide concentrations from Table 8.3. These assumed values are 1 x 10 $^{-10}$ curie/m³ for mixed fission products from the LWR fuel, 3 x 10 $^{-10}$ curie/m³ for the mixed fission products from FBR fuel, and 4 x 10 $^{-13}$ curie/m³ for the mixed actinides from either type of fuel.

Maximum site boundary distances dictated by the routine release of radionuclides to the atmosphere were estimated by assuming average annual concentration parameters that prevail in the direction northeast of CRNL (Fig. 8.5). Figure 8.8 compares this concentration parameter for the northeast direction at ORNL with corresponding parameters that have been estimated for the Hanford, 50, NRTS, 51 and Savannah River Sites. 49 The dashed curve labeled "I" shows the concentration parameter for iodine at ORNL that would result if the iodine were depleted from the plume with a deposition velocity of 0.04 m/sec. 52 The ORNL, Hanford, and NRTS data presented in Fig. 8.8 are based on meteorological calculations averaged over annual-weather conditions, but they are known to be reasonable based on long-term environmental monitoring studies. The Savannah River data are derived from results of air sampling studies for 131 made at the site boundary over a period of one year. The Savannah River data reflect the depletion of iodine in the plume.

Table 8.6 presents estimates of the site boundary distances and resultant average annual concentrations of the various species of radio-nuclides that would be dictated by routine releases from conceptual IWR and FBR plants sited at ORNL. These estimates assume that the plume is not depleted by deposition, fallout, and washout. Table 8.6 also gives estimates of the average annual concentrations of radionuclides at the site boundaries of the NFS, 16 MFRP, 12 and BNFP plants. These latter results were taken from the Safety Analysis Reports for the three plants; thus the assumptions made in the calculations are not necessarily the same

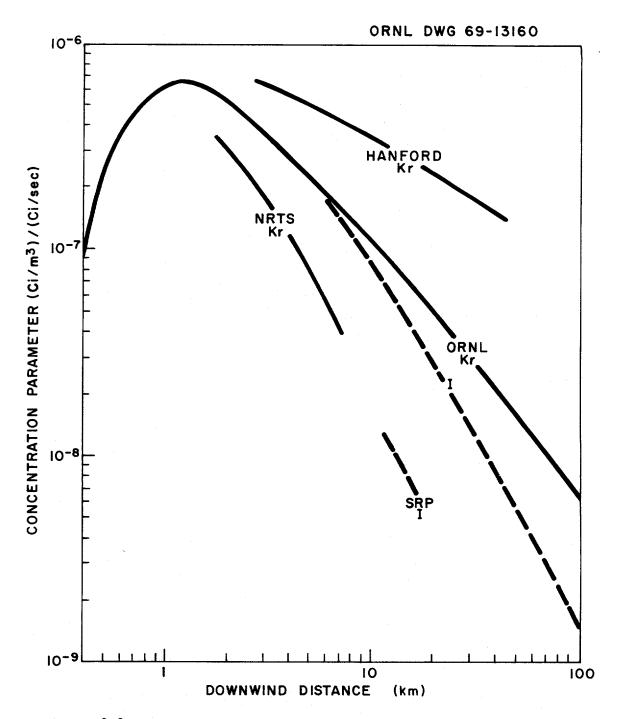


Fig. 8.8. Effect of Downwind Distance on the Average Annual Downwind Ground Concentration per Unit Emission Rate from a 100-m-tall Stack.

Table 8.6. Fraction of Maximum Permissible Average Annual Air Concentrations Resulting from the Routine Release of Radionuclides at the Site Boundaries of Existing, Proposed, and Conceptual Private Industrial Fuel Processing Plants

(260 days of operation per year)

		Fuel C	Fuel Characteristics	38	Distance	Average Annual	Fraction	of 1/3 x(100FR2	() Concentrati	Fraction of 1/3 x(100FR20) Concentrations at Site Boundary ^{3,b}	ya,b
Plant	Plant Gapacity (metric tons/day)	Burnup (Mwd/ton)	Specific Power (Mw/ton)	Decay Period (days)	to Site Boundary (km)	Aeolian Dilutign ($\sec(m^3)$)	85 _{Kr} -133 _{Ke}	Э,н	129 <u>1</u> -131	Fission Product Solids	Actinide Solids
NFS	1	20,000	32	150	1.5	2.2 × 10 ⁻⁷	0.23	0.002 (18,000)	0.47 (3.1)	0.0007 ^c (~1)	, ,
MFRP		43,800	30	160	0.6-3	1.1 x 10 ⁻⁷	0.12 (3,300,000)	0,005	0.23 (3.1)	<0,0005 (<2.2)	<0.11 (<0.63)
BNFP	5.8	35,000	01	160	٥	5.7 x 10 ⁻⁸	0.24 (1.4 × 10 ⁷)	0,02 (600,000)	0.27 (21)	0.003	0.017
LWR	T	33,000	30	150	9.0>	6.3 x 10 ⁻⁷	0.58 (2.9 × 10 ⁶)	0.054 (180,000)	0.15 (0.56)	0.003	0.021 (0.43)
LWR	9	33,000	93	150	0.5-6	1.8 × 10 ⁻⁷	(1.7×10^7)	0.093	0.25 (3.4)	0.002 (41)	0.018 (1.3)
LWR	38	33,000	30	150	5-29	3.0 × 10 ⁻⁸	1.0 × 10 ⁸)	0.093	0.25 (20)	0.001	0.009
FBR	Ħ	33,000	85	30	9*0>	6.3 × 10 ⁻⁷	0.92 (4.6 × 10 ⁶)	0.073 (240,000)	0.52 (3.6)	0.0003 (4.5)	0.008
FBR	9	33,000	28	30	1.5-10	1.1 × 10 ⁻⁷	(2.8×10^7)	0,079 (1,450,000)	0.56 (22)	0,0001	0.003
FBR	36	33,000	28	30	7-42	1.9 × 10-8	(1.7×10^8)	0.079	0.56 (130)	0.0001	0.003

a factor of 700, resulting in a reference concentration of 1.4 \times 10⁻¹³. The 10CFR20 value for ¹³1 was reduced by a factor of 700, resulting in a reference concentration of 3 \times 10⁻¹⁵. The 10CFR20 value for ¹²⁹1 was reduced by a factor of 7000, resulting in a reference concentration of 3 \times 10⁻¹⁵. ^aThe reference values selected are one-third of the concentrations found in 100FR20, Appendix B, Table II, Column 1. They are 1 x 10⁻⁷, 7 x 10⁻⁸, 1 x 10⁻¹⁰, 3 x 10⁻¹⁰, and all x 10⁻¹³ for ⁸⁵Kr - ¹³³Xe, ³H, mixed Lims fission products, mixed FBR fission products, and mixed actinides respectively. The 100FR20 value for ¹³¹L was reduced by

^bRelease rates, in curies/year, are given in parentheses.

as those employed for the present analysis of conceptual plants. The comparisons are of value in that they reflect the range of results that can be obtained through the use of various assumptions and computational techniques, as well as point out differences that may exist in meteorological conditions from site to site.

The large site boundary distances that are estimated for plants of high capacity provide incentive for removal of a larger fraction of the noble gases and iodine than was assumed in Sect. 8.2.1. This will be considered further in Sect. 8.4 after estimates are presented of the site boundary distances that are dictated by upper limit accidents.

8.3 Accidental Releases of Radioactive Materials

Fuel processing plants utilize three barriers for the confinement of radioactive materials. Accidents may cause the primary barrier to fail and, in turn, radioactive gas, liquid, or aerosol (usually under pressure) to be discharged to the second barrier. The first confinement barrier consists of the process vessels, the associated interconnecting piping, and the highly efficient vessel off-gas train. The second barrier is the thick concrete cell wall, which is designed to provide radiation shielding and to limit the effect of the maximum explosion in a process vessel within the cell to minor leakage of air or gas to the third barrier. barrier, an industrial building, surrounds all penetrations in the cell walls. Under normal conditions, outside air is drawn into the building through (1) a roughing filter, (2) a check valve and another roughing filter to the cells, and (3) a ventilation duct (where it mixes with the effluent from the off-gas train) and HEPA or deep-bed filter to blowers, which exhaust to a stack. Normally a portion of the ventilation air from the building does not pass through the cells but flows directly, through a suitable restriction, to the upstream side of the filters. In an accident situation, in which one or more cells may become pressurized, this latter flow tends to maintain the building at a negative pressure with respect to the environment. Glove-box facilities have three barriers the box, the laboratory, and the building - which have comparable confinement potential to the vessel, cell, and building. Mobile materials in storage canals are confined by a container, the water, and a building.

Potentially, liquid waste management facilities also have three barriers of confinement — the tank, a vault, and a building. In present practice, however, massive failure of the tank (such as by a hydrogen-air explosion in the vapor space), resulting in significant pressurization of the vault, is not considered credible because of the assumed reliability of preventive measures; therefore, the third barrier (a backup floor pan and a building) may not be considered necessary. By making the more pessimistic assumption that a hydrogen-air explosion in a waste tank is credible, it is assumed in this study that either the waste tank or the vault (which is possibly vented through a large pipe to other vaults or to cells of the processing plant) is designed to contain the explosion (a maximum of ~100 psig in the vapor space of the tank), resulting in only minor leakage that is confined to a building and routed through a filtered ventilation system.

The following basic assumptions were made for the purpose of assessing the effects of credible accidents in fuel reprocessing plants:

- (1) The secondary containment barrier (cell, vault, water in the storage pool, and ventilation-filter system) and the building can, and will be, designed to maintain their confinement potential following exposure to any credible internal forces.
- (2) Process and confinement systems can, and will be, designed in such a manner that exposure to credible external events or forces (loss of power, earthquake, tornado, flood, hurricane, impaction by moving vehicles, etc., but not including acts of war) will not impair the ability to shut down the plant safely and maintain safe shutdown conditions.

The following sections will describe more detailed assumptions that have been made with respect to the properties of fuel reprocessing plants and waste management facilities, estimates of the fractional release of radioactive materials resulting from accidents, a model for the assessment of downwind consequences of a release, and implications of the estimated dose rates as a function of distance downwind.

8.3.1 Assumed Properties of Fuel Reprocessing Plants

Properties of fuel reprocessing plants as a function of capacity (see Table 8.7) have been assumed for the purpose of estimating the fractional release of radioactive materials in the event of an accident. With a few exceptions, the containment and confinement features that were selected represent either present or only moderate extensions of current technology. Future large-capacity plants will, undoubtedly, have many properties different from those selected; however, it is assumed that the important derived numbers (i.e., the quantities of radioactive materials released in accidents) will remain unchanged or decrease with advancing technology.

The assumed properties are for central plants processing spent fuels from light water (IWR) or fast breeder (FBR) reactors employing unit operations of chop-leach, solvent extraction, and ion exchange. A schematic drawing of the type of plant that is assumed is shown in Fig. 8.9. It is assumed that spent fuels are stored prior to processing in water-filled canals. High-level wastes are assumed to be either pot-calcined immediately and stored in water-filled canals for two years prior to shipment or stored for two years in an acid solution and then calcined prior to shipment. Low-level wastes are assumed to be discharged predominantly to the atmosphere. Intermediate-level wastes (spent solvent, resins, etc.) are assumed to be fixed in asphalt, polyethylene, or concrete; and hulls are assumed to be stored in vaults in relatively small containers.

Process Equipment. - It is assumed that the concentrations of fuel (U + Pu) in aqueous solutions in the head ends of the LWR and FBR plants are 0.3 and 0.1 metric ton/m³ respectively. The volume of fuel solution in a single vessel was kept relatively small, 3 to 30 m³, by assuming that the relative processing rate will have increased by a factor of 3 (because of more continuous equipment) by the time that 18-metric ton/day LWR plants or 9-metric ton/day FBR plants are built, and that the 36-metric ton/day plants for LWR and FBR fuels consist, respectively, of two 18-metric ton/day and four 9-metric ton/day independent modules. Multiple tanks of these assumed sizes, in separate compartments to prevent interaction in the event of an accident, would be employed if additional capacity is needed for head-end equipment (dissolver, and accountability and solvent extrac-

Table 8.7. Assumed Properties of Reprocessing Plants and Waste Storage Facilities

		Fuel Proce	essing Rate	(metric t	ons/day)a	
		LWR Fuel			FBR Fuel	
	1	6	36	1	6	36
Processing plant						
Total dissolver solution, m ³ /day	3.33	20	120	10	60	360
No. of independent lines	1	1	2	1	1	4
Relative processing rate/line	1	1	3	1	3	3
Max. head-end vessel capacity, m ³	3.33	20	20	10	20	30
Total cell capacity/line, m ³	2333	14,000	14,000	7000	14,000	21,000
No. of cells/line	7	11/1	14	7	14	1)4
Cell size, m ³	333	1000	1000	1000	1000	1500
Cell ventilation rate, m ³ /min	66.7	200	200	200	200	300
Total ventilation rate/line, m ³ /min	700	4200	4200	2100	4200	6300
Ventilation train ^b	F,M	F,M	F,M	F,A,M	F,A,M	F,A,M
Total off-gas flow rate	28	85	255	2.0	4.0	5/1
Off-gas train ^b	S,T,F	S,T,F	S,T,F	S,I,F	S,I,F	S,I,F
Interim ^C liquid waste (acid) storage facility						
Tank volume (80% filled), m ³	812	3785	3785	990	3785	3785
No. tanks required for 2-year accumulation	2	3	10	2	3	13
Off-gas flow rate/tank, m ³ /min	6.1	28	28	7.4	28	28
Off-gas train ^b	C,F	C,F	C,F	C,F	C,F	C,F
Vault ventilation rate, m ³ /min	6.1	56	224	7.4	56	280
Ventilation train ^b	C,F,M	'C,F,M	C,F,M	C,F,M	C,F,M	C,F,M
Interim ^C waste solids storage canal						
Length for 14.6-m width, m	5.8	35	210	7.1	42	250
Ventilation rate, m ³ /min	170	1000	61.00	210	1200	7300
Ventilation train ^b	C,F	C,F	C,F	C,F	C,F	C,F

^aA 1.0-metric ton/day plant processes 260 metric tons of uranium + plutonium per year.

bS = caustic scrubber; 90% removal of iodine.

T = silver tower; 99% removal of iodine.

A = activated charcoal filter; 99% removal of iodine.

M = metal mesh or silica gel; 99.9% removal of Te, Cs, and Ru.

I = high-efficiency iodine removal unite; iodine DF of 107.

C = steam condenser; discharges air at 100°F and 100% relative humidity.

F = either reliably-protected HEPA or deep bed filter. Normal effluent = 0.0012 mg/m 3 . Accident effluent = 0.02 mg/m 3 .

^CTwo years.

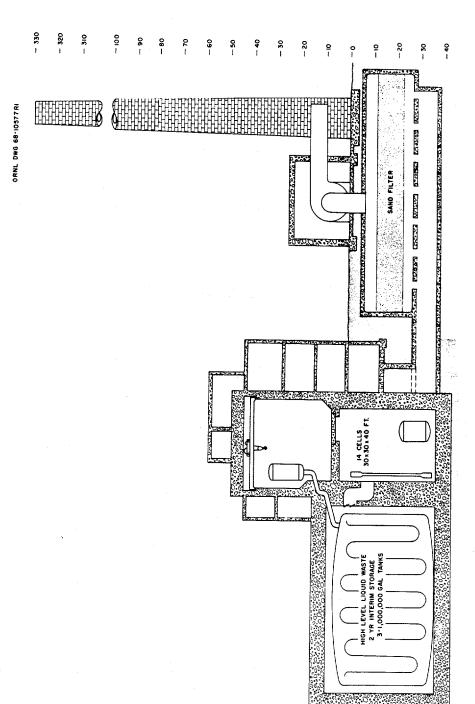


Fig. 8.9. Spent-Fuel Processing Plant with a Capacity of 6 Metric Tons/Day.

tion feed tanks) or plutonium storage (tanks of the assumed maximum size packed with borosilicate-glass raschig rings containing solution at a plutonium concentration of 0.25 metric ton/m³).

Process Cells. - Process cells are assumed to have reinforced (1 to 2% steel) concrete outer walls that are approximately 5 ft thick, reinforced concrete partition walls between cells that are approximately 2 ft thick, and volumes 50 to 100 times greater than the maximum vessel. Such cells, roughly 25 to 35 ft cubes that have secured roof plugs, could withstand a sustained pressure of 30 to 50 psig or the detonation of 25 to 50 lb of TNT at their geometric centers without rupture. It is assumed that 7 cells are used in small plants; however, more compartments (i.e., 14 cells per process line) are used in plants having capacities greater than 6 metric tons/day.

Vessel Off-Gas System. - Present technology, with a trend toward relatively lower off-gas flow rates per unit of plant capacity, is assumed for LWR plants. It is also assumed that FBR plants will be designed to minimize the vessel off-gas flow rate to approximately 70 cfm in a l-metric ton/day plant and that this flow will vary directly with plant capacity but inversely with relative processing rate. The off-gas is assumed to pass through a train (wet scrubber, solid halogen absorber, and filter) to effect partial removal of iodine, semivolatile fission products, and particulates and to discharge to the ventilation system on the upstream side of the ventilation filter. It is assumed that the wet scrubber serves to retain about 93% of the iodine in a relatively nondispersible form and that the off-gas train for FBR plants will include devices which will provide a cumulative DF of 10⁷ for iodine.

Ventilation System. - The ventilation air exhaust is assumed to consist of the air flow from the cells (at 0.2 air change per min) plus an additional 50% that flows directly from the building (third containment barrier). This stream is filtered, passed through metal mesh or silica gel for 99.5% removal of ruthenium vapors, and finally exhausted to the atmosphere through a 100-m-tall stack. In addition, the FBR plant is assumed to be equipped with activated charcoal filters for 9% removal of iodine. The filter system is assumed to be composed of either a sand

filter or roughing and HEPA filters with equivalent reliability and integrity. Independent process lines are assumed to have independent ventilation systems.

Facility for Interim Storage of Liquid Wastes. - The interim liquid waste storage facility is assumed to provide for two-year storage of acid waste (at a concentration of 0.01 gal per Mwd of burnup) consistent with a maximum tank size (80% filled) of 1,000,000 gal and at least 30% spare tankage. The off-gas stream - 1000 cfm for a 1,000,000-gal tank is assumed to pass, first, through a condenser (which would condense and recycle the distillate to the tank in the event of loss of coolant), then through a filter, and finally be discharged to the ventilation system for the vault. The latter ventilation system collects the small purge flow from each tank vault (plus the combined off-gas from all tanks) and discharges it through a backup condenser, filter, and ruthenium removal device to a 100-m stack. The tanks and/or the vault are assumed to be designed to withstand a hydrogen-air explosion (an internal pressure of ~100 psi) without rupture, possibly by venting to other tanks or vaults. The tanks, vaults, and ventilation system are assumed to be designed to withstand the effects of the maximum earthquake.

Canal for Interim Storage of Waste Solids. - The canal for interim storage of waste solids (i.e., calcined waste) is assumed to provide for a two-year accumulation of 6-in.-diam by 10-ft-long pots, each containing fission products from 14,100 Mwd of burnup at an average solids concentration of 1.0×10^{-14} ft³/Mwd. The pots are assumed to be covered with at least 20 ft of water. The ventilation system for the canal and building provides 12 air changes per hour to minimize fog formation. The ventilation system is assumed to be exhausted through a dehumidifier and HEPA filters at the roof of the building.

8.3.2 Analytical Models and Mechanisms of Accidental Release

Mechanisms that tend to negate the primary confinement barrier (process vessels, associated piping, and the efficient, low-flow off-gas system) have the potential of releasing radioactive materials to the atmosphere through the ventilation system. The following sections will describe

models for predicting the fractional release, discuss dispersive mechanisms, and present estimates of the fractional release to the atmosphere from upper limit accidents.

The designs of models for the release of radioactive materials depend on whether the material is released to the ventilation system as a gas (or vapor) or as an aerosol.

Gas or Vapor. - Certain of the fission products (the noble gases. halogens, and semivolatiles) may escape from the primary containment barrier in gaseous form. The release to the environment from such sources is relatively easy to predict; it is the fractional release from the vessel mitigated by the removal efficiency of the devices in the ventilation train. The noble-gas fission products, dominated by 85 Kr and 133Xe, may be released essentially quantitatively from process vessels. Devices for partial removal of noble-gas fission products are not used in present commercial reprocessing plants, but several types of devices have been proposed for this application. 51,53 The halogens, dominated by 131I and ^{129}I , may be volatilized from process operations as I_2 , HI, or organic iodides. Since these compounds have high vapor pressures at room temperature, they are not appreciably removed by filtration. Usually, activated charcoal filters may be relied upon to remove 99% of the iodine from a ventilation stream, especially if most of the iodine is in the form of ${\rm I}_{2}$ or HI (the typical forms released from most process operations).

Certain other fission products, notably (in approximate order of importance) Ru, Cs, Te, Tc, and Se, may be classed as semivolatiles since gases or vapors of these elements may result from certain abnormal process operations. The oxides of Se and Tc are completely volatilized at temperatures in the vicinity of 200°C, while the normal oxides of Ru, Cs, and Te require temperatures generally greater than 750°C. 54 Under highly oxidizing conditions in acid solutions, ruthenium may form the tetroxide, which has a boiling temperature of approximately 80°C. A slight excess of KMnO₁ in an acid uranyl nitrate solution at 80°C will result in the volatilization of 70 to 80% of the contained ruthenium in 5 to 10 min. 55 For this reason, highly oxidizing conditions are avoided in present fuel

reprocessing plants. Evaporation and complete boildown of a nitric acid solution of fission products will result in the volatilization of 10 to 20% of the ruthenium. 56,57 Once airborne, the vapor tends to rapidly deposit on metal surfaces and decompose to the relatively nonvolatile dioxide. For this reason, a "bucket of Brillo" (i.e., a tank packed with stainless steel mesh) has been found to be effective for removing ruthenium from off-gas and ventilation streams at the Savannah River Plant. Silica gel absorbers, operating at about 70°C, were found to remove 99.6% of the ruthenium from waste calciner off-gas at Idaho. 58

Radioactive Aerosols. - The aerosol that would be dispersed in cell air by an accident would consist of a dispersion of a radioactive solution, solid particles, or smoke. The physical properties of aerosols are such that they effectively restrict the escape of radioactive particles to the environment. This is seen commonly in practice since, through the use of appropriate deentrainment mechanisms, the condensate from the evaporation of a radioactive solution may be made to contain only $10^{-1/4}$ to 10^{-6} of the activity of the solution. Gravitational settling serves to limit the maximum aerosol concentration; we have been able to demonstrate this through an approximate correlation of the solution concentration in air or vapor arising from cooling towers, evaporators, and airsparged vessels. This correlation is shown in Fig. 8.10.

In order to properly describe the release of aerosols from a cell, we must be able to ascribe removal efficiencies to filters and to cracks in cell walls. For superficial velocities less than approximately 0.15 ft/sec, it has been found that an aerosol formed by vigorous mixing of a solution with air is metastable and has a concentration of the order of 10 mg/m³. This metastable concentration is approximately equivalent to fog, which has a concentration of approximately 10 mg/m³ and a particle size of approximately 10 μ . For orientational purposes, a 1-in./hr rain with a mass mean particle size of 3000 μ has a concentration of 1000 mg/m³. At ORNL, ⁵⁹ the particle size distribution of the metastable aerosol in a ventilation stream downstream from the source has consistently been found to have the particle size distribution shown in Fig. 8.11. Another piece of relevant information reported by Garner ⁶⁰ is that the weight distribu-

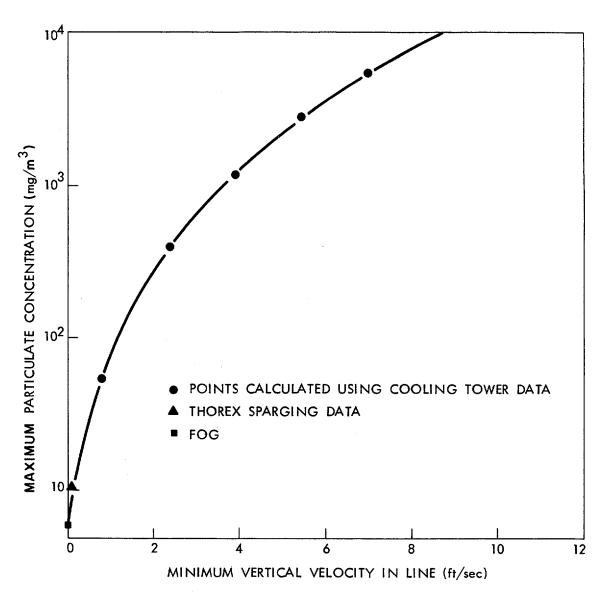


Fig. 8.10. Effect of Minimum Superficial Velocity in an Off-Gas Line on the Concentration of Liquid Solution Particles Resulting from Vigorous Mixing of a Solution with Air. Density of solution, 1 g/cc.

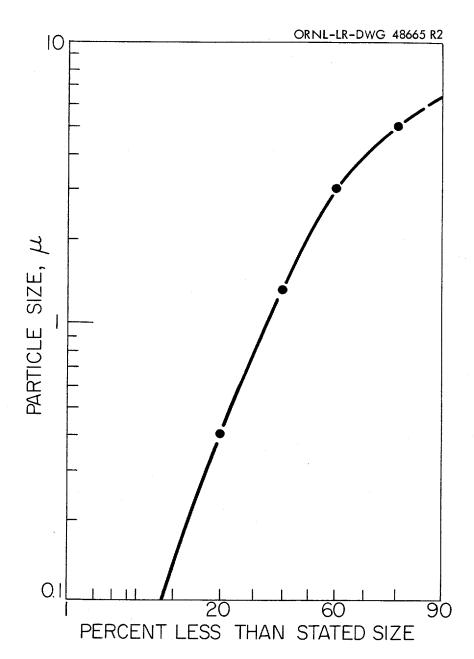


Fig. 8.11. The Particle Size Distribution of a Stable Aerosol Which Has Encountered Several Changes of Direction in a Pipeline.

tion of particles smaller than 10 to 20 μ will be fairly constant, even if there is gross entrainment of larger droplets. The knowledge that this distribution is fairly constant and constitutes approximately 10 mg/m³ may be used to estimate the approximate concentration of particles smaller than a given size, even in an air stream which is very concentrated with liquid droplets. Practically, it is possible to assign efficiencies to an absolute filter and calculate the effluent concentration.

In evaluating the concentration of aerosols in air which leaks from a cell, it is considered that the aerosol must follow many small tortuous paths in its escape through 5 ft of concrete. The evaporator deentrainment studies by Walsh and Schlea⁶¹ indicate that a single right-angle impingement will reduce a liquid aerosol concentration to 10 mg/m³ or less. Fine heavy-element dust would be reduced to the order of 1 mg/m³, and the concentration of smoke in leaked air would probably be no more than approximately 100 mg/m³. These numbers are primarily of use in estimating the radiation dose to plant operating personnel. Essentially all of the material that escapes from the cells through cracks during a period of temporary pressurization would be routed through the filtered cell ventilation system.

Junge⁶² and Friedlander⁶³ have observed that the particle size distribution of airborne aerosols is remarkably constant or "self-preserving." Small particles tend to agglomerate rapidly by Brownian motion, while large particles are removed by impingement or sedimentation. Friedlander suggests that a quasi-stationary state exists such that the rate at which matter enters a differential size is equal to the rate at which matter is lost by sedimentation. It has also been determined that the stable concentration of small particles (less than about 3 μ in diameter), because of agglomeration, is consistently less than a few grams per cubic meter after the aerosol has been permitted to "age" for a few seconds or minutes. ⁶⁴⁻⁶⁶ Friedlander has proposed the following formulation for the differential concentration of particles in a metastable aerosol as a function of size:

$$dn = k'r^{a-1} dr , (6)$$

where

n = number of particles per unit volume,

k' = a constant,

r = radius of the particle.

a = a constant with a value of ~-1 to -1.5.

By converting to a mass concentration and integrating from r=0 to r=r, the concentration of particles with less than a given diameter is:

$$C(\langle D) = k_{\rho}D^{3+a} , \qquad (7)$$

where

C(<D) = mass concentration of particles with diameter less than D, mg/m^3 ,

 ρ = density of the agglomerate, g/cm³,

 $D = diameter of the agglomerate, \mu$,

k = another constant.

It has been found that agglomerates, even of dense particles, have a density of approximately 1 $g/cm^3.67$

A related expression may be derived using the largely substantiated expression for the rate of agglomeration of an aerosol containing particles of a single size: 64

$$\frac{dn}{dt} = Kn^2 \quad , \tag{8}$$

where

t = time since beginning of agglomeration,

K = agglomeration coefficient. 64

 $\approx 9 \times 10^{-11}/D \text{ cm}^3/\text{sec}$ for $D \leq 0.3 \mu$,

 $\approx 3 \times 10^{-10}$ cm³/sec for D $\geqslant 0.3 \mu$.

From this expression, the initial concentration, Co, of particles having a diameter Do and a half-life of $\rm t_{1/2}$ is:

$$Co = \frac{\pi \operatorname{Do}^{3}_{\rho}}{6Kt_{1/2}} . \tag{9}$$

Assuming a half-life of 10 min and converting to appropriate units, this expression becomes:

$$Co = 9700 \rho Do^{4}$$
 $Do \leq 0.3$ (10)

$$Co = 2900 \rho Do^3$$
 $Co \ge 0.3$ (11)

These expressions for aerosol concentration are compared with experimental data for a wide variety of heavily concentrated and turbulent aerosols (smoke, flyash, DOP, etc. in air, and water droplets in air and steam) in Fig. 8.12. Expressions (10) and (11), for concentrations of monodispersed aerosols with a half-life of 10 min, provide a practical upper bound for the concentrations of solid particles in air. A better description of aerosols containing liquid particles, is provided by expression (7) when a is approximately equal to -2.

Based on the maximum concentration of particulates as a function of particle size (see Fig. 8.12) and assuming that the efficiency of deepbed sand or HEPA filters is 100% for particles 0.3 u in diameter, the predicted concentration of particles in the effluent from absolute filters is 0.02 mg/m3. Cheever determined experimentally that the maximum concentration of plutonium particles in the effluent from a 30-in.-deep sand filter, occurring at the optimum superficial velocity for a penetration of 4.8 ft/min, was 0.02 mg/m³ (ref. 68). This experiment was performed under conditions that are very unlikely to occur in accident situations; the filter influent concentration was 100 mg/m³, and the count-mean particle size was only 0.07 u because the aerosol had aged for only a few seconds. Cheever also found that an HEPA filter removed an additional 99% of the particles in the effluent from the sand filter. From these data and known characteristics of filter systems, it is assumed that filter effluent concentrations of 0.02 mg/m³ or less are attainable in practice, regardless of the mass concentration of the influent.

The release of radioactive material through the cell ventilation system by a mechanism that generates aerosols is estimated to be as follows:

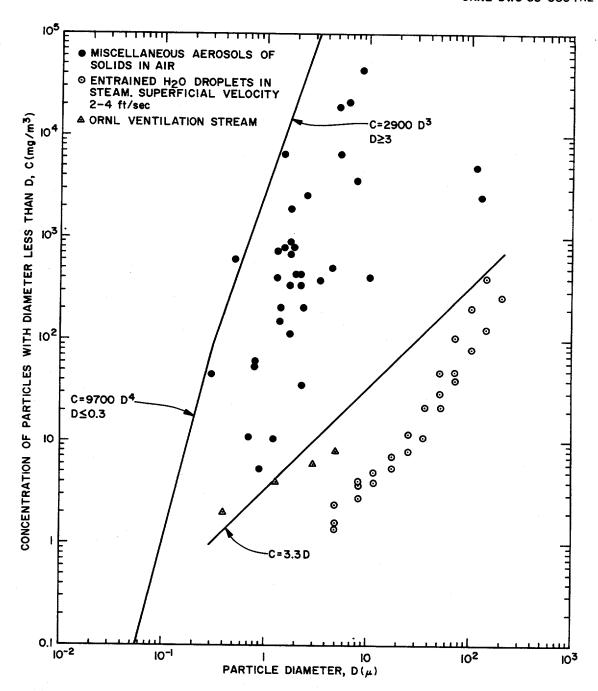


Fig. 8.12. Mass Concentration of Solid and Liquid Particles in Aerosols.

$$R = C \frac{D_{p}}{D_{f}} \left[F_{v}^{t} + V_{c} \frac{F_{v}}{F_{c}} \right] , \qquad (12)$$

where

R = the quantity of a component (in one metric ton of fuel) that is released to the atmosphere,

C = mass concentration of particles in the filter effluent = 2×10^{-8} kg/m³.

 $\mathbf{D}_{\mathbf{p}}$ = concentration of the component in the particles, weight fraction,

 $D_{\mathbf{f}}$ = concentration of the component in the fuel, weight fraction,

 $F_v = \text{total flow rate in ventilation system, } m^3/\text{min,}$

 F_c = flow rate from the cell in which the aerosol has been generated, m^3/min ,

t = duration of the source term for aerosol generation, min,

 V_c = volume of the air in the cell (evaluated at one atmosphere of pressure) following the dispersion of aerosol, m^3 .

When the aerosol is formed essentially instantaneously, as in an explosion, the rate of release to the atmosphere will decrease exponentially with a mean life of F_c/V_c (which is assumed to be 5 min).

Dispersive Mechanisms. - Mechanisms for the dispersion of gases and aerosols in cells include chemical explosions, fires, nuclear excursions, and leakage. Some properties of explosions relative to the containment potential of cells are shown in Table 8.8. All of the quantities of the limiting explosive materials are very large as compared with their credible inventories in a process vessel. The allowable quantities are even larger if the cells are vented to another confinement zone of large volume (i.e., the cell-canyon concept used at Hanford and planned for MFRP). It is assumed to be incredible that the cell would first fill with hydrogen or solvent vapor and then explode. The flow rate of cell ventilation air is sufficient to dilute any radiolytic H₂-O₂.

The most serious fires in a fuel processing plant would be those involving plutonium, that is, solvent or ion exchange resin loaded with

Table 8.8. Estimated Properties of Explosions That Could Occur at the Center of a 10-m³ Cell^a with 5-ft-thick Reinforced (1 to 2% Steel) Cell Walls Without Rupture

Total Energy Release (Btu)	Pressure at Cell Wall (psig)
54,000	≤100
1,500,000	50
1,500,000	50
~1,500,000	50
~1,500,000	50
9,100,000	~0.7
	Release (Btu) 54,000 1,500,000 1,500,000 ~1,500,000

^aInside dimensions.

 $^{^{\}rm b}$ Maximum burst of 10^{16} fissions/liter in a tank containing 30,000 liters of solution at a temperature of 85°F.

plutonium. Purex-type solvent will burn at the rate of about 1 in. of depth per hour and generate approximately 20,000 Btu/hr per ft² of burning surface. Experience in gloved enclosures has shown that fires covering the entire area of the floor of the enclosure tend to self-extinguish in a matter of minutes because of depletion of the oxygen. This has been true even in well-ventilated enclosures because the pressure increases to several inches of water and reverses the flow through the intake. It has been observed that ion exchange resin loaded with plutonium nitrate can ignite spontaneously at about 120°C and burn (in the absence of air), liberating about 540 Btu/lb.

Experience has shown that the initial burst resulting from a super prompt-critical nuclear excursion in a solution is limited to a maximum of 10¹⁹ fissions per m³ of solution.⁶⁹ At this fission density, the void coefficient caused by the generation of radiolytic gas (~1.4 m³ of gas, at STP, per m³ of solution) is sufficient to override the effect of high reactivity addition rates. Assuming that the temperature of the solution is 85°F (the yield would be lower if the temperature were higher), this burst would increase the temperature to boiling. If the solution is not rendered permanently subcritical by the initial or succeeding bursts or by ejection of solution, it may possibly boil to dryness. The dried solids, if not subcritical because of low density and lack of moderation, probably would be dispersed by one last burst.

Assuming that all of the solution in an equilateral cylinder with a volume V_T is involved in a nuclear excursion, the upper limit yield of the initial burst (and probably the most powerful burst, resulting in the generation of a radiolytic gas void fraction, at STP, of 1.4) is $10^{19} \, V_T$ fissions. When boiling begins, the steam void coefficient (2.3 x $10^{-17} \, \mathrm{m}^3/\mathrm{fission}$) (ref. 70) would limit individual bursts to approximately 6.1 x $10^{16} \, \mathrm{fissions/m}^3$. Since the height of the solution in the tank is $1.08 \, \mathrm{V}^{1/3}$, and the bubble rise rate is about 12 m/min, the period between bursts is approximately 0.09 $\mathrm{V}^{1/3} \, \mathrm{min}$. The total time required for boildown of the solution (assuming 2.1 x $10^6 \, \mathrm{Btu/ton}$) is about 52 $\mathrm{V}^{1/3} \, \mathrm{min}$.

The sudden generation of radiolytic gas or steam in the solution would cause an inertial force to be exerted against the walls of the

tank. An overestimate of the maximum amount of work that can be done in deforming the vessel, taking no credit for free expansion into the vapor space of the tank, may be calculated assuming that the liquid and gas expand reversibly against the plastic flow pressure of the vessel. Assuming a gas void fraction (at STP) of 1.4 per burst, no more than about 0.5% of the energy released in the burst could do pressure-volume work against a resisting pressure of 200 psig. An unrestrained cylindrical tank of characteristics assumed in this study could, theoretically, withstand repeated bursts without rupture. [The rupture strain of 304L stainless steel is 0.65 (ref. 71)].

8.3.3 <u>Method of Analysis of the Downwind Consequences of a Unit Release</u> of Radioactive Material

The method that has been selected for investigation of the environmental consequences of an accidental release of radioactive material from a fuel reprocessing plant consists, first, of the examination of a "unit" release of activity and, second, the application of the resulting data to actual releases which could be expected from the various credible accidents.

Two different mixtures of isotopes have been considered. These mixtures (listed in Table 8.3) simulate the fission product and actinide contents of typical spent LWR fuel and LMFBR core and blanket fuel mixtures which will actually be encountered. It is assumed that the LWR fuel has decayed for 150 days prior to processing and that the LMFBR material has decayed for 30 days.

For these mixtures, a "unit" release is defined to be the release of all materials that are associated with 1 kg of fuel; therefore, the fundamental calculations have been performed on this basis. Initially, no provision is made for differences in the chemical or physical behavior of the various isotopes, and it is assumed that all of the components in a unit release escape. However, as will be shown, it is possible to treat differences in behavior and thus account for variations in the release fraction due to filtration, chemical reactions, and other processes that affect some of the components but not others.

We have investigated both the external gamma dose and the external beta dose that result from direct exposure to the radiation flux originating in the plume and from the internal radiation dose received as a result of the inhalation of radioactive material by a receptor submerged in the plume. The calculation of both types of doses depends on a knowledge of the concentration of radioactive material in the plume as a function of time and space. The concentrations have been computed by using the "Gaussian Plume" formula 72 and by utilizing the source term and ground reflection correction described by Binford, Barish, and Kam. 73 source term is derived using the assumption that a unit quantity of radioactive material is released into the processing building, where it is instantly and uniformly mixed with the air in the building. It is further assumed that a constant fraction of the building volume is being discharged from the stack per unit time. These assumptions lead to the following expression for the concentration at the space point (x,y,z), relative to an origin of Cartesian coordinates at the stack orifice, and at time $\boldsymbol{\tau}$ after the release has occurred:

$$X(\mathbf{x}, \mathbf{y}, \mathbf{z}, \tau) = \frac{\alpha q e^{-\lambda \tau} \cdot e^{\alpha(\mathbf{x}/\mathbf{u} - \tau)}}{2\pi u \alpha_{\mathbf{y}} \alpha_{\mathbf{z}}} e^{-\mathbf{y}^2/2 \sigma_{\mathbf{y}}^2}$$

$$\times \left[e^{-\mathbf{z}^2/2 \sigma_{\mathbf{z}}^2} + e^{-(2\mathbf{h} + \mathbf{z})^2/2 \sigma_{\mathbf{z}}^2} \right], \quad \tau \ge \mathbf{x}/\mathbf{u}$$
(13)

where

 $X(x,y,z,\tau)$ = concentration, (curies/m³), q = initial release, (curies), u = wind speed in the x-direction, (m/min), x,y,z = space coordinates (m), $\sigma_y(x)$, $\sigma_z(x)$ = horizontal and vertical dispersion parameters, 72

respectively, (m), α = exhaust rate, (min⁻¹), λ = decay constant, (min⁻¹), h = effective stack height, (m), τ = time since release, (min).

Decay will be neglected for the mixtures under consideration so that λ is set equal to zero. Moreover, the value of the concentration at the plume center line (y=0) at ground level (z=-h) is of great interest. Under these conditions,

$$X(x,o,-h,\tau) = \frac{\alpha q e^{\alpha(x/u - \tau)}}{\pi^{u}\sigma_{y}\sigma_{z}} \cdot e^{-h^{2}/2\sigma_{z}^{2}}, \quad \tau \ge x/u$$

$$= 0, \quad \tau < x/u.$$
(14)

This expression is proportional to the inhalation dose rate at ground level at the plume center line and very nearly proportional to the external beta dose. The time integral from $\tau = 0$ to $\tau \to \infty$ is then proportional to the total dose. This integral,

$$X(x,o,-h,\tau) d_{\tau} = \frac{-h^2/2\sigma_z^2}{\pi^u \sigma_y \sigma_z} , \qquad (15)$$

is independent of α , the exhaust rate.

The dispersion parameters $\sigma_y(x)$ and $\sigma_z(x)$ are monotonic increasing functions of the downwind distance, x; however, they also vary with atmospheric stability. For a given value of x, the dispersion parameters decrease with increasing stability. It is, therefore, necessary to specify the degree of atmospheric stability in order to select the appropriate set of values for the parameters. For the purpose of investigating the external doses, two sets of atmospheric conditions have been utilized:

- (1) "Most Representative Conditions," where the wind speed has been chosen to be 100 m/min (3.73 mph) and slightly unstable (C) conditions are assumed to prevail.
- (2) "Inversion Conditions," where the wind speed is 50 m/min (1.86 mph), moderately stable (F) conditions prevail, and an inversion "lid" exists just above the stack orifice. To account for the latter, the vertical dispersion parameter is modified by being held constant once it reaches the value $\sigma_{\rm z}({\rm x}) = {\rm h/2.15}$ (see ref. 72).

Many other combinations of wind speed and stability conditions are possible; however, it is believed that these two are reasonably typical, cover most of the likely situations that may arise, and permit valid interpolation to other cases which lie in between.

The inhalation doses have been computed on a somewhat more comprehensive basis. As suggested above, many different combinations of wind speed and stability conditions are possible. Thus the inhalation doses have been computed for each of six different stability conditions, the results have been plotted on a single graph, and the envelope of the curves thus obtained have been utilized to estimate the inhalation dose to be expected at each ground level point downwind on the plume center line.

In all cases, it is assumed that the effective stack height, h, is 100 m. (The effect of stack height on ground-level concentration will be discussed in detail in a later section.) Finally, it should be pointed out that all of the doses computed below assume exposure of the receptor during the entire course of the accident.

External Beta and Gamma Doses. - These doses stem from direct exposure of the receptor to the radiation flux in the plume. Because of their short range, only the beta particles that originate in the vicinity of the receptor contribute to the dose. Hence, the dose rate may be assumed to be proportional to the concentration of beta emitters at the location of the receptor. The gamma dose, on the other hand, requires a space integration over the entire volume of the cloud in order to sum the photon flux incident on the receptor.

The computer program PLUME, ⁷³ which was originally developed in order to calculate internal iodine and external iodine and noble-gas doses following a reactor accident was used to perform these calculations. Input for the beta dose calculation is the average energy per disintegration, the equivalent number of curies, and a numerical constant to convert Mev/m³ into dose units. Input for the gamma calculation consists of the gammaemitting inventory, divided into nine energy groups, and the appropriate cross sections and buildup factor parameters. The results are displayed in Figs. 8.13 and 8.14. Note that, in these cases, there is no physical

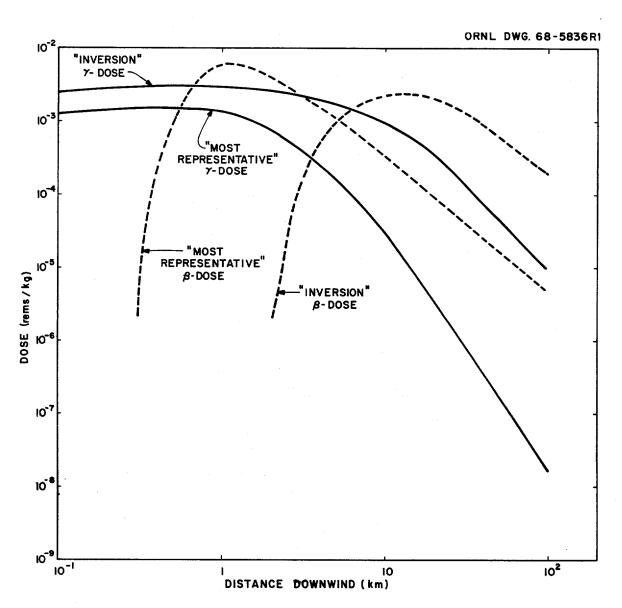


Fig. 8.13. External Radiation Dose due to All Fission Products Released in Reprocessing 1 kg of LWR Fuel.

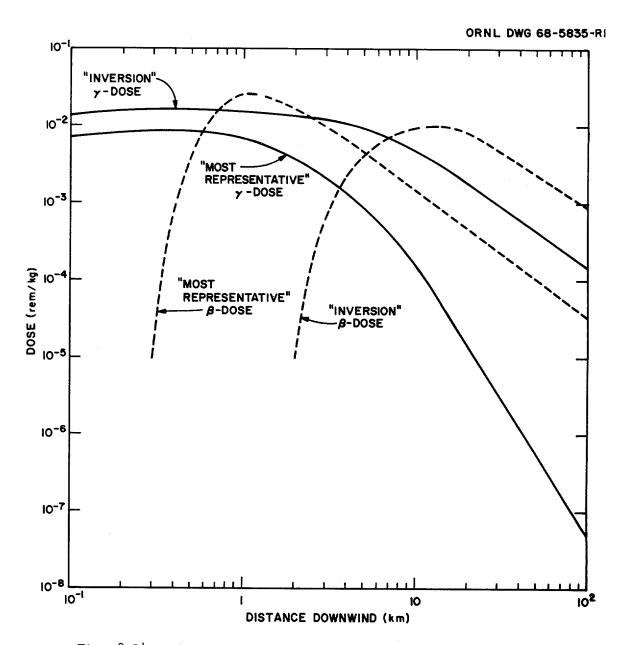


Fig. 8.14. External Radiation Dose due to All Fission Products Released in Reprocessing 1 kg of LMFBR Mixed Core and Blankets.

separation of the components and that all of the isotopes present are assumed to behave similarly.

Inhalation Dose Calculations. - These radiation doses result from inhalation of the contaminated air in the plume and from subsequent deposition of radioactive material in the various organs of the body. The rate of intake of radioactive material is proportional to the breathing rate and to the concentration of the radioactive material at the location of the receptor. The total intake is simply the time integral of the product of these two quantities. For the purpose of these calculations, it will be assumed that the receptor is located at ground level on the center line of the plume and that the exposure lasts for the duration of the accident so that the intake rate is integrated over infinite time. If the breathing rate is assumed to be constant, the total intake is:

$$I = \frac{-h^2/2\sigma_z^2}{\pi^u \sigma_y \sigma_z} \text{ curies }, \qquad (16)$$

where β is the breathing rate in m³/min and the other symbols are as defined previously. For this study, β has been taken to be 2.08 x 10 $^{-2}$ m³/min. The quantity I/q is the total amount of activity inhaled per curie of originally released material.

It should be noted that the spatial variation of the inhalation dose is independent of the amount of released material, the wind speed, and the breathing rate. Thus, for a given wind speed, breathing rate, and quantity of material released, the expression

$$9 = \frac{e^{-h^2/2\sigma_z^2}}{\sigma_y \sigma_z}$$
 (17)

is the same function of x, regardless of the amount of material that is released. This function, normalized to unity at its maximum value, has been plotted in Fig. 8.15 for each of the six stability conditions. An envelope enclosing the six curves has been drawn; this envelope permits estimations of the dose at each point downwind by using a knowledge of the dose at any given point.

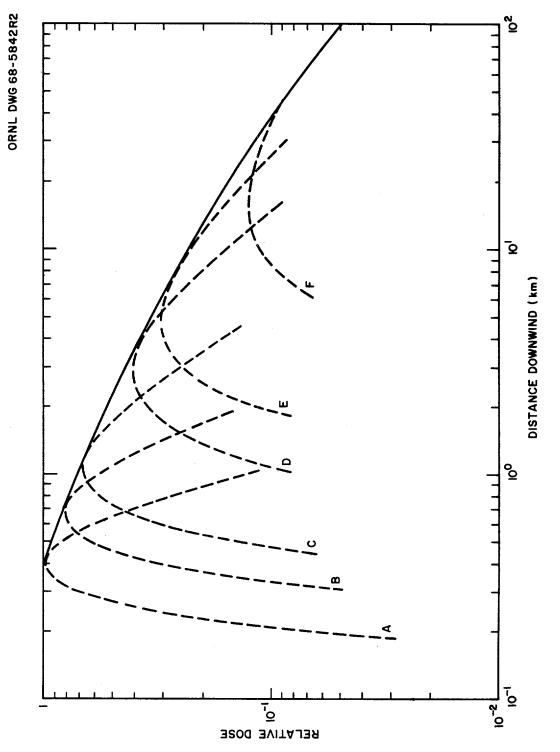


Fig. 8.15. Generalized Curve for Estimating Inhalation Doses as a Function of Distance Downwind (h = 100 m). The fraction of released material that is inhaled by a person μ 00 m downwind is estimated to be $\mu.3 \times 10^{-9}$

The dose at 400 m under A (extremely unstable) conditions is the maximum and has been chosen for reference. At a wind speed of 100 m/min, the total intake following the release described above is 4.3 x 10⁻³ uc per curie released. This factor has been utilized as input data for a computer program INREM, ⁷⁴ which, given the quantity of radioactive material inhaled, computes the dose to the most important organs as a function of time after inhalation. The program takes into consideration uptake by the various organs, effective half-life, and the age of the receptor, which, for this study, was chosen to be 20 years.

The INREM Computer Code. - The rate of intake of radioactivity is the primary radioactivity input for calculating the cumulative dose equivalents by the INREM Code. These estimates of dose are compiled for the various body organs from inhalation or ingestion of radioactivity programmed as continuous or intermittent intakes as a function of age. The parameters in the dose equations change as a function of time as the person ages during the time of intake or during the period of interest (which may be longer than the period of intake). This code, as currently dimensioned, has the capacity to handle 110 radionuclides and 11 body organs. The model, programmed for all organs except the gastrointestinal (GI) tract, is written as follows:

$$D_{in}(t_{1}, t_{2}, t_{b}) = 51 \int_{t_{1}}^{t_{2}} I_{i}[t - t_{b}), t] f_{in}(t - t_{b}) \left\{ \int_{t}^{t_{2}} \frac{\varepsilon_{in}(s - t_{b})}{m_{n}(s - t_{b})} \right\}$$

$$\times \exp[-\int_{t_{1}}^{s_{1}} \lambda_{in}(\gamma) d\gamma] ds dt , \qquad (18)$$

where

 $D_{in}(t_1, t_2, t_b)$ = cumulative dose equivalent (rems) received during the time interval t_1 to t_2 from the ith radionuclide in the nth organ, resulting from intake during this time interval by an individual born at t_b ,

t₁ = time (days) of initial intake relative to time of release (t = 0 at time of release),

t₂ = time (days) at end of period of interest relative to time of release,

th = time (days) of birth relative to time of release,

t = time (days) after release,

s = time after intake relative to time of release,

 $I_{i}(t) = intake (\mu c/day)$ of ith radionuclide at t,

 $m_n(t) = mass (g)$ of the nth organ at t,

f_{in}(t) = fractional absorption (dimensionless) of the ith radionuclide in the nth organ at t,

 $\epsilon_{in}(t)$ = effective absorbed energy (Mev) of the ith radionuclide in the nth organ at t, and

 $\lambda_{in}(t)$ = effective elimination constant (day⁻¹) of the ith radionuclide in the nth organ at t.

The variables t_1 , t_2 , t_b , t, and s are measured relative to release, whereas the variables I(t), $m_n(t)$, $f_{in}(t)$, $\varepsilon_{in}(t)$, and $\lambda_{in}(t)$ are functions of the age of the individual. The code uses Eq. (18) for ingestion of contaminated food and water, or inhalation of contaminated air, and calculates the cumulative doses to all organs except the GI tract.

When the age-dependent cumulative dose equivalents to the GI tract are to be calculated, the ${\rm (MPC)}_a$ or ${\rm (MPC)}_w$ is used in the following way:

$$D_{iyz}(t_1, t_2, t_b) = \frac{O.3}{7I_y'(MPC)_{iyz}} \int_{t_1}^{t_2} I_{i}[(t - t_b), t] m_s/m_\ell(t - t_b) \varepsilon_{i\ell}(t - t_b)/\varepsilon_{is}$$

$$\times f_{i\ell}(t - t_b)/f_{is} dt$$
 , (19)

where

 $D_{iyz}(t_1,t_2,t_b)$ = cumulative dose, equivalent (rems) to a critical segment of the GI tract, received during the time interval t_1 to t_2 from inhalation (y = 1) or ingestion (y = 2) of the soluble (z = 1) or insoluble (z = 2) form of the ith radionuclide for an intake during this time interval by a person born at t_b ,

 $I_y' = intake (cc/day) of air (y = 1) or water (y = 2),$

 $(MPC)_{\mbox{iyz}}$ = maximum permissible concentration ($\mu c/cc$) of the ith radionuclide in air (y = 1) or water (y = 2), where the ith radionuclide is soluble (z = 1) or insoluble (z = 2),

t₁ = time (days) of initial intake relative to time of release,

t₂ = time (days) at end of period of interest relative to time of release,

 t_{b} = time of birth relative to time of release,

 $I_{i}(t)$ = intake ($\mu c/day$) of the ith radionuclide at t,

s = standard man index,

 ℓ = age index,

 m_{ℓ} = mass (g) of the critical segment of the GI tract for the ℓ th age group,

 $\varepsilon_{i\ell}$ = effective absorbed energy (Mev) of the ith radio-nuclide in the critical segment of the GI tract in the ℓ th age group,

 $f_{i\ell}$ = fractional intake of the ith radionuclide reaching the critical segment of the GI tract in the ℓ th age group.

Calculations were made with the INREM code to determine the dose commitment for the first year following inhalation (which, in this model, is the highest annual dose commitment) and also the dose commitment for a period of 50 years following the intake. The complete output data from

INREM have been reproduced in Tables 8.9 through 8.12. The results at 400 m were then utilized to obtain the doses at each point downwind by means of the generalized curve of Fig. 8.15.

In order to allow for differences in chemical and physical behavior of the various isotopes, the isotopes may be divided roughly into categories, depending upon their volatilities, as follows:

- 1. Volatile fission products: noble gases, halogens, tritium
- 2. Semivolatile fission products: Ru, Te, Cs, Tc, Se
- 3. Nonvolatile fission products: all other fission products
- 4. Nonvolatile actinides: plutonium and transplutonic elements
 The dose commitment to the various organs, as well as to the whole body,
 by these categories is given in Table 8.13.

<u>Deposition</u>. - In all of the foregoing calculations, it has been tacitly assumed that there is no depletion of the plume by deposition, fallout, or rainout. Consequently, the results thus far obtained are conservative in that some depletion of the plume due to these mechanisms will occur. On the other hand, the deposition of relatively large quantities of an extremely toxic substance, such as ⁹⁰Sr or plutonium, on the ground in and around a highly populated area may give rise to a serious hazard. ⁶⁹

Criticality Accidents. - One possible cause of a serious accident in a fuel reprocessing plant is inadvertent criticality that results in a nuclear excursion. Aside from damaging mechanical effects, such an accident would augment the inventory of fission products to an extent depending on the number of fissions taking place during the excursion.

In order to assess the additional radiation doses that would result from such an incident, a "unit" nuclear excursion of 3.7 x 10^{18} fissions has been investigated. The iodine isotopes and the noble gases and their daughters were considered to be of primary importance. The internal dose due to iodine and the external dose due to both iodine and noble gases plus their daughters, have been calculated for both the "most representative" and "inversion" conditions, using the PLUME computer program. 73

Table 8.9. Internal Dose at $400~\mathrm{m}$ Downwind Following the Release of 1 kg of LWR Fuel Inhalation dose commitment (in rems) integrated over 1 year Intake period of 1-day duration; begins at age 20

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LIVER			NO DATA			NO DATA	3.034E-02	1.660E-02	1.102E-08		į	10136101	3.2955-05	1.675F-09	8.113E-08	6.170E-06	3.360E-05	1.669E-04	2.095E-06	3.265E-04	3.861E-07	NO DATA	8.623E=02	3.3415-03	2.366E-07	6.714E-06	3.2586-03	1.324E 00	2.689E-05	3.893E-06	4.656F-03	40-201-1 4-508E-04	6.365E-04	NO DATA	2.127E-08	2.799E-01	3.068E-02	4.444E-02	1.965E-01		2.668E 00	6.525E-01	5.417E 00
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TOTAL BODY	4.703E-03	4.506E-04	6.587E-03	3.077E-02	Z. 035E-04	1.9575.03	20-27-50 B	2.974F-09	2.924F-04	4.9725-03		5.269F-07				1.267E-04		4. 5055-00	•			4. 985F -02			1.320E-05		3. 978E-04				1.125E-05		1.971E-04	1-8346-05	1.310E-08	4.482E-02	4.913E-03	7.116E-03	7.239E-02	3.186E-03	Γ.	Ξ.	6.552E-01
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Table 8.9 (Continued)

TRACT	SOLUBL	.1436-0	.744E-0	. 86 6E-0	. 86 6E-0	. 86 6E-0	.320E-0	.889E-0	. 50 7E-0	.353E-0	.300E-0	094E-0	862E-0	0525-0	304E-0	00 E-0	0-3096	056E-0	0-3E69	485E-0	238E-C	847E-0	522E-0	897E-0	1.9716-07	764E-0	890E-0	204E-0	766E-0	124E-0	5755-0	4975-0	013600	25.7F=0	492E-3	045E-0	447E-0	407E-0	0-3E06	1056-0	0-3966	68 SE-0	224E-0	634E-0	40 7E-0	524E-0	7.269E-01
6.1.	OLVBLE	.286E-0	.690E-0	.320E-C	• 320E-0	+320E-0	. 740E-0	. 40 7E-0	.507E-0	•353E-0	.843E-0	.047E-0	.242E-0	.701E-0	.4435-0	.707E-0	.568E-0	.542E-0	.693E-0	.971E-0	. 807E-C	.236E-0	.695E-0	0-36 56 ·	9.857E-09	.535E-0	448E-0	. 81 5E-0	. 510E-0	994E-0	. 575E-0	0-325E.	0-3000	A A FE - O	30 6E-0	.224E-0	.447E-0	.725E-0	.269E-0	.255E-0	- 99 6E-C	.237E-0	.482E-0	.028E-0	.725E-0	.770E-0	2.505E-01
LUNGS	SOLUB	.791E-0	.311E-0	.433E-0	.433E-0	.433E-0	.968E-0	.428E-0	.314E-0	.423E-0	.485E-0	.866E-0	.898E	.267E-0	.964E-0	.334E-0	.241E-0	.590E-0	.601E-0	.969E-0	.271E-0	.219E-0	.612E-0	.225E-0	.915E-0	.522E-0	.329E-0	. 772E-0	.715E-0	.517E-0	•005E-0	0 144E 0	0-1000	574F-0	980E-0	.532E-0	.180E-0	.519E-0	.481E-0	.135E 0	.243E-0	.801E-0	.234E-0	.682E-0	.520E 0	061E-0	1.2C4E 01
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Table 8.10. Internal Dose at 400 m Downwind Following the Release of 1 kg of LWR Fuel Inhalation dose commitment (in rems) integrated over 50 years Intake period of 1-day duration; begins at age 20

NUCLIDE LABEL TOTAL BODY BONK NUCLIDE THYRELD LIVER NO DATA
H-15 Fe - 10 F
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NUCLIDE LABEL 1074L H-3 303 3.700E SR-90A 308 4.774E SR-90A 901 4.506E SR-90C 902 6.606E Y-90 643 2.035E XH-103 88 2.035E RH-103 88 2.035E SR-125 159 1.0354E SR-125 162 1.0354E SR-127 170 9.420E TE-129 1.759 TE-129 1.7
NACCA-155 EU-155 PU-239

Table 8.10 (Continued)

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Table 8.11. Internal Dose at $\mu = 100 \, \mathrm{m}$ Downwind Following the Release of 1 kg of LMFBR Fuel

Inhalation dose commitment (in rems) integrated over 1 year Intake period of 1-day duration; begins at age 20

OVARIES NO DATA																	NO DATA			NO DATA		NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	MU DATA	AT AC	NO OATA	NO DATA	ATAG DA		244	ATAC ON	ATAG ON	NO DATA	NO DATA	0.0					
TESTES NO DATA	NO DATA	NO DATA				NO DATA					ATAC CM			NO DATA	ATAG ON			NO DATA		136E	3.737E-03	4-962E-05	1.714E-02	3.045E-05	7.5726-05	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA				ATAC CA						NO DATA					Š			NO DATA	2.1176-02
SPLEEN NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	1.753E-01	7.4745-02	NO DATA	NO DATA	NO DATA	NO DATA	4140	2.629F-05	NO DATA	1.566E-04	3.553E-03	4.571E-05	1.785E-02	3.1 79E-05	9-1746-05	NO DATA	NO DATA	8.751E-03	7.590E-04	2.863E-02	1.169E-04	NO DATA		NO DATA						NO DATA							NO DATA	NO DATA	3.101E-01				
K I DNEYS NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	2.922E-01	9.260E-02	6-463F-05	8-418F-09	1.446F-07	4-831F-02	20 1426-01	3.755E-05	9.695F-05	1.619E-04	NO DATA	NO DATA	NO DATA	6.184E-04	1.403E-02	1.804E-04	7.046E-02	1.2556-04	3.621E-04	NG DATA	NO DATA	4.154E-03	6.513E-04	1.318E-02	9.318E-05	NO DATA	3.30dE-02	1.446F-02	6.619E-03	2.342E-02	2.954E-07	2.141E-04	4.632E-06	9.866E-03	1.905=03	6 310E-03	2-448E-01	2.054F-01	1.912F 00	1.753E-01	3.391E 00	9.5396-02	9.002E 00
LIVER NO DATA	NO DATA	NO DATA	NO OATA	NO DATA	NO DATA	NO DATA	2.308E-01	8.523E-02	2.6995-05	5.999F-10	1.148F-08	NO DATA	N CN	9.4495-06	3.3825-05	2.001E-04	1.489E-09	2.714E-05	1.5335-05	7.275E-05	1.650E-03	2.123E-05	8.821E-03	1.585E-05	4.882E-05	NO DATA	NO DATA	1.174E-02	1.054E-03	3.435E-02	Z-878E-04	60-101c1-0	2.2016.00	2.496F-02	1.412F-02	1.653E-02	2.185E-07	4.495E-04	4.114E-06	7.933E-03	4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4 4	0.0285-00	3.282E-01	3.9615-01	1.025E 00	3.518E-01	1.165E 01	3.250E-01	1.791E 01
THYRGID NO DATA						NO DATA	NO CATA	NO DATA	NO DATA	2.073E-08	2.282E-05	1.411E-06	3.600E-05	9.017E-04	3.252E-05	5.179E-03	2.104E-05	4.389E-05	8.716E-01	9.725E-04	NO DATA						NO DATA						NO DATA							NO DATA		8.788E-01							
MUSCLE NO DATA	NO DATA	NO DATA	NO DATA	NO DATA		NO DATA					NO DATA			NO DATA		NO DATA										NO DATA	NO DATA	1.049E-02	0.944E-04	20-3636-05	1.3185-04	4 F 4 C C Z	ATAC ON	NO DATA	ALAC CA	4 4 4 6 6 6 4	4 T 4 C	ATAC ON	NO DATA			NO DATA	NO DATA	3.456E-02					
				3.125E-01					2	10	7		-	-	7.883E-0				1.416E-03							ջ	_		3.2995-04			1.0075-02		6.244E-02					4.293E-06	1.7135-02	60-10-0	7.0236.00	č	2,585E 00	1.839E 01	3.110E-01	1.121E 01	3.0776-01	5.036E 01
TOTAL BODY 4.984E-07	3.120E-02	2.553E-04	3.732E-03	2.083E-02	1.1566-04	3.466E-02	1.413E-01	5.060E-02	5.096E-06	1.110F-08	3.100E-09	5-7776-03	1.564F-02	5.737E-06	1.7526-05	6.393E-06	3.396E-06	5.885E-05	3.147E-04	2.158E-05	4.304E-04	9.544E-06	3.371E-03	9.817E-06	3.488E-05	1.553E-03	2.370E-06	6. 788E-03	90-3788-0 40-1788-0	70-1404.1	1.000E-02	8.420E-03	2.597F-01	3.099E-03	9.150E-04	5.019E-03	8.1395-08	4.587E-05	1.569E-06	2.436E=U3	101111111111111111111111111111111111111	1.7875-01	5.255F-02	6.342E-02	3.777E-01	2.501E-02	452E-	2.044E-02	2.094E 00
LABEL 303	38	106	206	903	43	84	65	67	7.7	78	62	88	97	68	114	125	159	191	162	163	169	170	176	177	191	187	761	327	3 6	27.0	122	227	238	237	246	247	251	255	328	207	200	2 2	281	282	283	_	2	en	
NUCL I DE H-3	SR-89	SR-90A	SR-90B	SR-90C	V-90	Y-91	ZR-95	NB-95	40-99	TC-99M	TC-99	RU-103	RU-106	RH-103H	AG-111	CD-115M	SB-124	SN-125	SB-125	TE-125M	TE-127M	TE-127	TE-129M	TE-129	TE-132	1-131	1-132	CS-134	13-130	0.011.01	1 4-140	CF-141	CE-144	PR-143	NO-147	PM-147	PH-149	SM-151	261-03	TB-140	MD-1230	P11-23A	PU-239	PU-240	PU-241	AM-241	CM-242	CM-244	TOTAL
. NO																																																	

Table 8.11 (Continued)

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TRACT		CUBI	36-(37E-()-30c)-30¢) OE-(76-(/3E-() BE-(9E-(-36	36-(12E-() - 36)-36!	4E-(E)-36 (-3E-	€.	, SE-(. 6E-C	75-0	1.6-0	<u>-</u>	4	9	36	2E-0	, i	9 6	9 6	, L	16-0	4E-0	0-30	- E	٦. و	9	ָה הייני	7 4	4 T	96-0	SE-0	4E-0	OE-0	5E-0	2E-0	1.488E-02	3E-0	2.455E 00
	:	2	-467E-	.430E-	. 31 4E-(. 31 4E-(. 31 4E-0	. 879E-(. 394E-(. 908E-C	. 20 BE-C	• 289E-C	. 572E-C	.921E-0	. 994E-C	. 90 6E-0	. 329E-0	. 1456-0	.0736-0	. 394E-0	, 51 9E-0	, 17 SE-0	432E-0	, 77 SE-0	,357E-0	3496-0	, 269E-0	41 ZE-0	314E-0	340E-0	634E-0	0-1000	951E-0	092F-0	451E-0	876E-0	850E-0	681E-0	4146-0	587E-0	32 6E -0	80 3F-0	594E-0	1865-0	• 69 €E-0	. 345E-0	.450E-0	. 81 7E-0	.377E-0	1.190E-02	•877E-0	1.3838 00
LUNGS		NSCLUBI	.758E-(.851E-(.945E-(.945E-(.945E-(.957E-(-271E-	. 760E C	.306E-(. 236E-(.497E-C	. 716E-C	. 661E-C	. 972E (434E-C	.318E-C	025E-0	103E-0	-204E-0	. 8885-0	.509E-0	. 222E-0	2486-0	. 516E-0	.683E-0	574E-0	227E-0	6235-0	1976-0	0-2010	2025-0	271E-0	128E-0	226E 0	880E-0	235E-0	148E-0	2795-0	0.000	718F-0	945E-0	029E-0	523E 0	330E 0	605E 0	209E-0	675E-0	1.100E 01	018E-0	3.682E 01
2		LUBL	DAT	DAT	DAT	DAT	DAT	DAT	NO DATA	DAT	DAT	DATA	90E	764E-1	DAT	DAT	NO DATA	DAT	DATA	83E	DATA	304E	DA D	DAT	OA.	DAT	DAT	1	NO DATA	UA! A	00000	010000	1.60AF-04	DATA	DAT	DAT	DAT	DAT	DAT	DAT	4 4	DAT	DAT	DAT	DAT	DAT	DAT	DAT	O DAT	NO DATA	O DAT	6.199E-03
		LABEL	303	38	106	902	606	4	89	60	67	77	78	79	80 I	16	68	114	125	159	191	162	163	169	170	176	2:	161	÷	267	307	- 6	221	222	227	238	237	246	247	251	0.00	262	272	330	280	281	282	283		~ .	m	
		NUCLIDE			8		g.	V-90	1	G	o.	66-0N	5	6	RU-103	2	2	=	= :	12	15	2	15	2	2	12	-	?;	n.	7:	2.5	10	BA-140	1	7	7	4	1:	* :	4 1	. E	3	9	2	23	23	2,4	24	4	CM-242	Š	TOTAL
		Š	-	~	en	4	'n	•	-	60	6	0	1	12	2	4	2	9	1	8	<u> </u>	ر 20	21	55	23	*	2	0 1	- 6	9 6	*	? ;	32	33	34	35	36	37	9	, c	? 7	. 4	4	44	45	46	47	8	64	٥ د	7	

Table 8.12. Internal Dose at 400 m Downwind Following the Release of 1 kg of LMFBR Fuel

Inhalation dose commitment (in rems) integrated over 50 years Intake period of 1-day duration; begins at age 20

ARIES	4	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	O A L A	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	DATA	NA IA	DATA	DATA		
8	2	물	Z	욷	물	물	2	욷	욷	웆	2	Ž	2	2	2	2	2	2	2	Ž	2	2	2	욷	웆	오	皇	웆	웆	2	2	29	2 2	2 2	2	2	2	물	욷	물	웆	₽!	2	2	2 :	₹ :	2 9	₹ 9	22	•	5
TESTES	NO ON A	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA		NO DATA				ND DATA			NO DATA	NO DATA	NO DATA	NO DATA	1.336E-04	3.737E-03	4.962E-05	1.714E-02	3.045E-05	7.572E-05	NO DATA	NO DATA	NO DATA		ND DATA	NO DATA	A L A C				NO DATA					NO DATA			NO DATA				NO DATA	2 1135.03	30-21 1117
SPLEEN NO DATA	4140	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	1.778E-01	7.478E-02	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	2.629E-05	NO DATA	1.566E-04	3.5546-03	4.571E-05	1.7858-02	3.1796-05	9.1746-05	NO DATA	NO DATA	9.276E-03	7.590E-04	3.091E-02	1.1695-04	NO CALA	NO DATA	NO DATA	NO DATA						NO DATA				NO CATA		NO DA A	NG DATA	3.754F=01	30171-01				
KIDNEYS		NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	NO DATA	2.963E-01	9.265E-02	6.493E-05	8.418E-09	1.4466-07	4.831E-02	2.162E-01	3.755E-05	9.695E-05	1.621E-04	NO DATA	NO DATA	NO DATA	6.184E-04	1.403E-02	1.804E-04	7.046E-02	1.2556-04	3.621E-04	NO DATA	NO DATA	4.161E-03	6.513E-04	1.321E-02	9.318E-05	3.369E-02	1.652E 00	1.446E-02	6.619E-03	4-850E-02	2.9546-07	6.605E-04	2.303E-05	2.251E-02	1.948E-03	2. (55E-05	2.902E 01	10 1910 1	10 2077 · 1	7000	6 190E 00	1.835E 00	9.803F 01	**
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Table 8.12 (Continued)

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Table 8.13. Summary of Maximum Inhalation Dose Commitments at 400 m Downwind Following the Release of 1 kg of LWR or Mixed LMFBR Fuel from a 100-m Stack

· · · · · · · · · · · · · · · · · · ·	Whole Body	Bone	Lungs	Liver	Thyroid
LWR	Fuel - First-Y	ear Dose C	ommitment		
Volatile fission products Semivolatile fission products	0.00000039 0.0695	- 0.0898	0.000018 2.56	- 0.120	0.0000136 0.000303
Nonvolatile fission products	0.242	3.74	4.83	1.38	-
Plutonium	0.129	5.78	1.48	0.552	_
Transplutonic elements	0.215	3.22	3.17	3.37	-
Total	0.655	12.8	12.0	5.42	0.000318
Ī	WR Fuel - Lifet	ime Dose C	ommitment		
Volatile fission products	0.00000039	_	0.0000317	_	0.0000136
Semivolatile fission	0.0709	0.0983	2.76	0.126	0.000304
products	0.000		.	- 41	
Nonvolatile fission	0.839	13.7	5.13	1.64	-
products Plutonium	3.51	151.	2.94	16.0	
Transplutonic elements	1.12	18.3	2.94 3.33	9.67	-
Total	5.54	183.	14.2	27.4	0.000318
LMFBR Cor	e-Blanket Fuel	- First-Ye	ar Dose Comm	itment	
Volatile fission products	0.00156		0.0123		0.872
Semivolatile fission products	0.0194	0.169	6.98	0.0578	0.00621
Nonvolatile fission products	0.610	8.29	10.4	2.68	-
Plutonium	0.672	30.1	7.68	2.87	_
Transplutonic elements	0.791	11.8	11.7	12.3	-
Total	2.09	50.4	36.8	17.9	0.879
LMFBR Con	re-Blanket Fuel	- Lifetim	e Dose Commi	tment	
Volatile fission products	0.00156	_	0.0123		0.873
Semivolatile fission products	0.0480	0.174	7.40	0.0604	0.00621
Nonvolatile fission products	1.01	15.4	10.9	3.14	-
Plutonium	18.6	806.	15.3	85.0	· _
Transplutonic elements	2.24	34.6	12.0	22.1	-
Total	21.9	856.	45.6	110.	0.879
	· · · · · · · · · · · · · · · · · · ·				

^aIn rems.

The input data for this calculation are presented in Table 8.14, and the results are given in Fig. 8.16.

In some instances, iodine may be retained on a charcoal filter; in such cases the thyroid dose shown in Fig. 8.16 would be reduced by a factor equal to one minus the filter efficiency. For very efficient filters, virtually all of the iodine would be retained; the whole-body dose would then be due only to the noble gases produced during the excursion and to those that result from the decay of the iodines trapped on the filter. The external gamma-ray dose delivered due to noble gases alone is shown in Fig. 8.17.

Validity of These Calculations. - The foregoing methods for estimating the downwind radiation doses following a nuclear accident have been developed using the "Gaussian Plume" model. 72,75 Implicit in this derivation are the assumptions that the degree of atmospheric stability, the wind speed, and the wind direction remain unchanged during the entire course of the incident.

Although the results have, in most cases, been extrapolated to a distance of 100 km from the stack, it is extremely doubtful whether this model is valid for distances of more than 20 or 30 km. At a speed of 100 m/min, it would require 17 hr for the plume to extend for a distance of 100 km. However, it is almost certain that variations of the weather conditions, both with time and distance, would occur.

Moreover, the model also assumes flat, featureless terrain and does not take into account the various topographical features such as hills, valleys, and lakes. No provision is made for the presence of buildings and other structures, which may affect the behavior of the effluent either because of proximity to the emitting source or because of modification of the behavior of the plume in the vicinity of the receptor. Items such as these must be handled on an individual basis, and, at present, there seems to be no obvious way of generalizing the results of these effects.

In all cases, it has been assumed that the release takes place at an elevation of 100 m. This assumption produces somewhat lower ground-level concentrations than would a similar release that occurs at ground level.

Table 8.14. Source Terms for Criticality Accident (based on 3.7 x 10^{18} fissions)

	Isotope	Yield	$\lambda (sec^{-1})$	q (curies)
	131 _T	0.029	9.96 x 10 ⁻⁷	2.9
	132 _T	0.044	8.02 x 10 ⁻⁵	352.9
	133 _I	0.065	9.25 x 10 ⁻⁶	60.1
	134 _I	0.076	2.20 x 10 ⁻¹ 4	1,672.0
	1351	0.059	2.89 x 10 ⁻⁵	170.5
	83m _{Kr}	0.0048	1.01 x 10 ⁻⁴	48.5
	85m _{Kr}	0.015	4.41 x 10 ⁻⁵	66.2
	87 _{Kr}	0.027	1.48 x 10 ⁻⁴	399.6
	88 _{Kr}	0.037	6.95 x 10 ⁻⁵	257.2
I	89 _{Kr}	0.046	3.63 x 10 ⁻³	16,698.0
7-	$133m_{Xe}$	0.0016	3.49×10^{-6}	0.56
	133 _{Xe}	0.065	1.52 x 10 ⁻⁶	9.9
	135m _{Xe}	0.018	7.40×10^{-4}	1,332.0
	135 _{Xe}	0.062	2.11×10^{-5}	130.8
	138 _{Xe}	0.055	6.79 x 10 ⁻⁴	3,734.5
	88 _{Rb}	Same as 88 Kr		257.2
II	89 _{Rb}	Same as ⁸⁹ Kr		16,698.0
	188 _{Cs}	Same as ¹³⁸ Cs		3,734.5
	133 _{Xe}	Same as 133I		60.1
III	$135 \mathrm{m_{Xe}}$	30% of ¹³⁵ I		51.2
	135 _{Xe}	70% of ¹³⁵ I		119.3

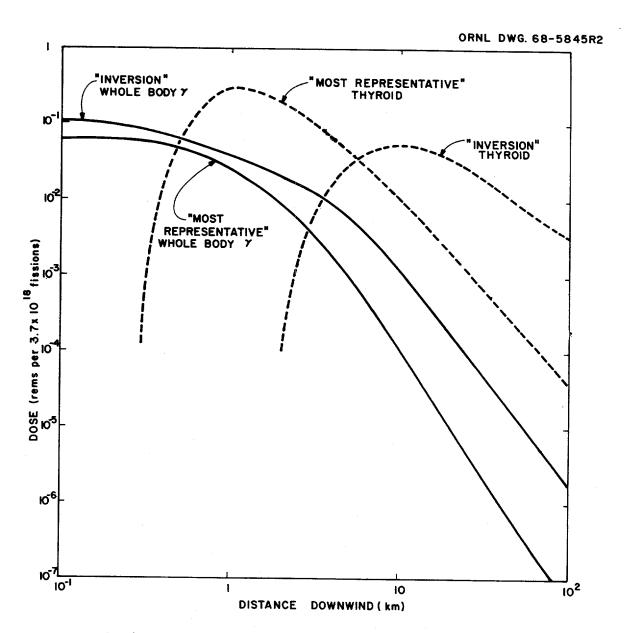


Fig. 8.16. Radiation Dose due to Volatile Fission Products Produced During a Nuclear Excursion (Based on 3.7 x 10^{18} Fissions).

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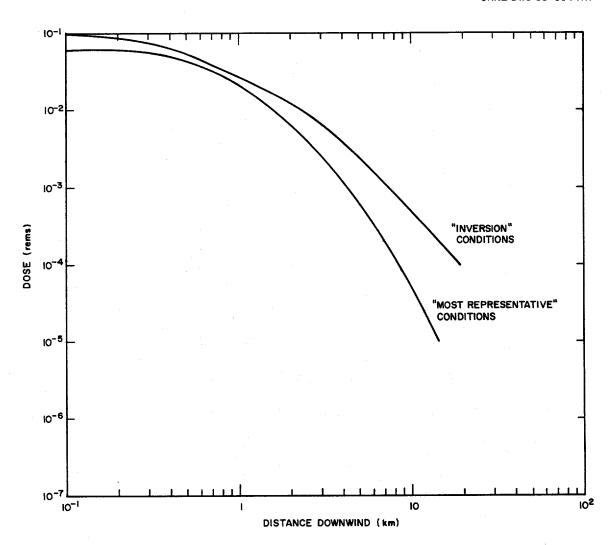


Fig. 8.17. Whole-Body External Gamma-Ray Dose due to Noble Gases Produced During a Nuclear Excursion (Based on 3.7 x 10^{18} Fissions). (Note: In this case it is assumed that all of the iodine isotopes are retained on filters.)

However, except for extremely stable meteorological conditions, the difference is quite small once the peak ground concentration produced by the elevated release has been passed. For example, under C-conditions, the ground concentration from a release taking place at an elevation of 100 m reaches 75% of that from a similar ground-level release at a distance of 1.2 km from the point of release.

The wind speeds used in these calculations have been chosen to be 100 m/min and 50 m/min because this range of wind speed is reasonably characteristic of many locations. However, as can be seen from Eq. (16), the dose is inversely proportional to the wind speed.

Finally, it should be pointed out that all of the doses calculated are those which are delivered at ground level on the plume center line. To obtain off-center-line ground-level doses, it is necessary to multiply the results by the quantity

$$e^{-y^2/2\sigma_y^2}$$
,

where y is the distance (in meters) normal to the plume center line at which the dose is required, and σ_y (in meters) is the horizontal dispersion parameter. For convenience, values of σ_y and σ_z for the various stability conditions are shown in Figs. 8.18 and 8.19.

Despite the obvious shortcomings of the procedures outlined, it is believed that they will, at least, produce order-of-magnitude results. These procedures will permit the development of sufficient "feel" for the magnitude of the various credible accidents so that the problem of siting can be approached in a quantitative manner.

8.3.4 Downwind Consequences of Upper Limit Accidents

Upper limit accidents were determined using the assumed properties of fuel reprocessing plants (Sect. 8.3.1) and models and mechanisms described in Sect. 8.3.2 such that the release of noble gases, "fresh" fission products, iodine, semivolatile fission products, nonvolatile fission products, and plutonium is maximized. The computed fractional releases from the most significant accidents are summarized in Table 8.15.

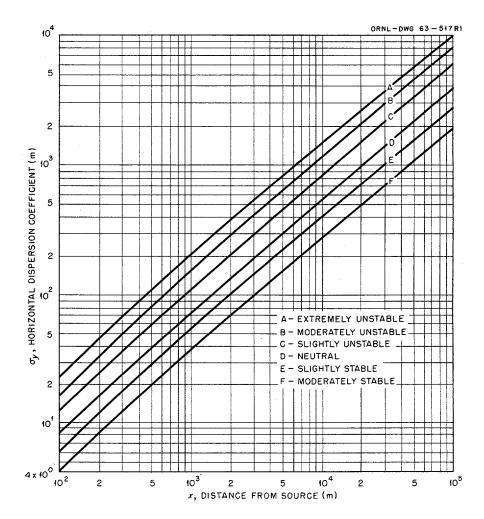


Fig. 8.18. Horizontal Dispersion Parameter as a Function of Distance Downwind.

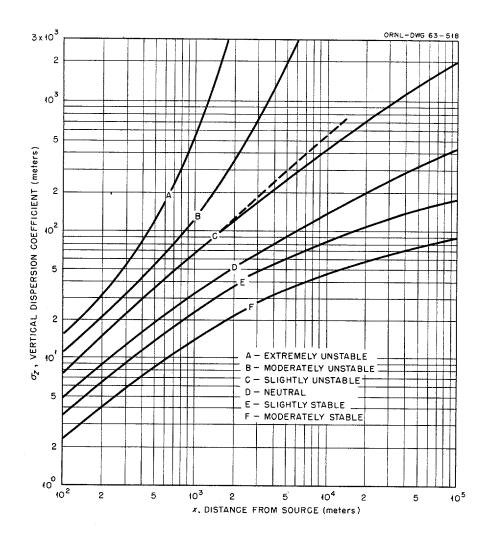


Fig. 8.19. Vertical Dispersion Parameter as a Function of Distance Downwind.

Table 8.15. Accidental Releases from Fuel Reprocessing Plants as a Function of Capacity

		Release (kg from Plant	of Fuel ^a Unles s of Capacity (s Otherwise Ind metric tons/day	dicated) 7) of:	
	LWR Fuel	Reprocessing P	lant	FBR Fuel	Reprocessing	Plant
Accident	1	6	36	1	6	36
Nuclear Excursion in Head End						
Duration, min	78	140	140	110	1740	160
No. of fissions	2.7×10^{20}	1.6 x 10 ²¹	1.6 x 10 ²¹	8.0×10^{20}	1.6 x 10 ²¹	2.4 x 10 ²¹
Noble gas, %	100	100	100	100	100	100
Iodine, %	30	30	30	1.0	1.0	1.0
Volatile fission products	300	2000	2000	10	20	30
Semivolatile fission products	1	6	6	1	2	3
Nonvolatile fission products	0.00043	0.0046	0.0046	0.00082	0.0020	0.0035
Transplutonic elements	0.00043	0.0046	0.0046	0.00082	0.0020	0.0035
Pu (head end)	0.00043	0.0046	0.0046	0.00082	0.0020	0.0035
Pu (Pu storage tank) ^b	0.035	0.37	0.37	0.018	م. المبله	0.075
Noble-Gas Release						
85 _{Kr +} 133 _{Xe} , curies	70,000	420,000	2,500,000	350,000	2,100,000	13,000,000
Halogen Release						
131 _I	780	4700	14,000	7.8	47	70
129 _I	36,000	220,000	660,000	360	2200	3300
Semivolatile Release						
Semivolatile fission products	1	6	6	1	2	3
Release of Nonvolatiles						
Semivolatile fission products	0.00075	0.0045	0.0045	0.0018	0.0037	0.0055
Nonvolatile fission products	0.00075	0.0045	0.0045	0.0018	0.0037	0.0055
Transplutonic elements	0.00075	0.0045	0.0045	0.0018	0.0037	0.0055
Plutonium Release						
Plutonium	0.045	0.27	0.27	0.016	0.032	0.048

^aThe release of a component of the fuel is the product of these numbers and the concentration of that component in a kilogram of average LWR or LMFBR fuel.

^bThe nuclear excursion in the Pu storage tank is estimated to have the same yield and duration, but would release only "fresh" fission products and plutonium.

Site boundaries dictated by the upper limit accidents were estimated assuming that the maximum acceptable annual dose commitments resulting from exposure to the cloud or inhalation at the site boundary are those recommended by the NCRP for annual occupational exposure. These emergency dose commitments are compared with those of lOCFR100, and Isochem land requirements study, and an ORNL study involving peacetime applications of nuclear explosives in Table 8.16. The assumed acceptable dose commitments have been employed only for reference purposes, but are believed to be reasonable in view of the very low probability of occurrence of the assumed upper limit accidents.

The maximum site boundaries (Table 8.17) for all LWR plants and the l-metric ton/day FBR plant are determined by the whole-body dose resulting from the release of volatile "fresh" fission products from a nuclear excursion (30% and 1% release of iodines from LWR and FBR plants, respectively, plus 100% release of the noble gases). Site boundaries for the larger FBR plants are determined by the thyroid dose resulting from a silver tower explosion, which is assumed to release 0.1% of the equilibrium inventory of iodine. In Table 8.18, the total dose commitments resulting from various upper limit accidents at the accident-dictated site boundaries of these conceptual plants are compared with estimated dose commitments at the site boundaries of the NFS, MFRP, and BNFP plants.

Noble Gases. - In plants that will partially remove the noble gases from off-gas streams, the upper limit accident involving these gases is considered to involve the complete release of the contents of a storage vessel that contains a 7-day accumulation of krypton and xenon. A release of approximately 6,400,000 curies of ⁸⁵Kr plus ¹³³Xe is required to cause a maximum (at 400 m) downwind whole-body dose of 5 rems. This quantity represents the total accumulation of these gases over 890, 148, and 25 days in LWR plants with capacities of 1, 6, and 36 metric tons/day, respectively, and the total accumulation over 680, 115, and 3 days in FBR plants with capacities of 1, 6, and 36 metric tons/day respectively. The release of the 7-day accumulation of ⁸⁵Kr and ¹³³Xe in a 36-metric ton/day FBR plant would result in a whole-body dose of greater than 5 rems within distances of about 2.3 km.

Table 8.16. Comparison of Assumed Maximum Dose Commitments for Individuals in the General Population as a Result of Upper Limit Accidents with Those Given in 10CFR100, an Isochem Land Requirement Study, and a Study for Excavation of a Sea-Level Canal with Nuclear Explosives

	Thi	s Study	10CFR100	Isochem Study	Nuclear Ex	cavation Study ^d
	Maximum Annual Dose Commitment (rems/year ^a)	Approximate Total Dose Commitment (rems/50 years ^b)	Maximum Total Dose Commitment (rems ^C)	Maximum Total Dose Commitment (rems/lifetime)	Maximum Annual Dose Commitment (rems/year)	Maximum Total Dose Commitment (rems/70 years)
Whole body	5	50	25	25	3	10
Red bone marrow	5				3	10
Head and trunk	5					
Gonads	5				3	10
Lens of eyes	. 5				8	15
Skin	30			300	15	30
Thyroid	30	30	300	300	15	30
Bone	30	500		300	15	30
Hands, forearms, feet, and ankles	75				38	7 5
Other single organs	15	90 (liver) 18 (lung)		150	8	15

^aThese data are maximum permissible annual doses for occupational exposure as recommended by NCRP.

bThese data represent the approximate 50-year dose commitment resulting from a single intake of mixed spent reactor fuel such that the maximum annual (first-year) dose commitments do not exceed those given in the first column.

^C100FR100 provides reference values of total whole-body and thyroid dose (incurred during passage of the radioactive cloud) for use in the evaluation of reactor sites with respect to potential reactor accidents of exceedingly low probability of occurrence and low risk of public exposure to radiation.

d These data are proposed maximum acceptable dose commitments for use in planning for the construction of a sea-level canal with nuclear explosives. They are considered applicable to special radiation protection problems in which an assessment of risk vs benefit would dictate greater annual dose commitments than those recommended by the ICRP, FRC, and IAEA.

Table 8.17. Site Boundaries (Distance from the 100-m Stack) Determined by the Maximum Upper Limit Accidents in a Spent-Fuel Processing Plant $^{\rm a}$

	Dista	ance to Site Cap	Boundary (km) for Repr ic tons/day)	rocessing Pi	lants of
		LWR Fuel			FBR Fuel	
Accident	1	6	36	1	6	36
Nuclear excursion	٢٠١١ (٥٠	2.0	2.0	1.2	2.0	2.8
Release of:						
Noble gases Halogens Semivolatiles Nonvolatiles Plutonium	(1.1) ^b (0.2) ^b (17) ^b (0.04) ^b (0.9) ^b	(6.6) ^b (1.0) ^b 0.14 (0.23) ^b (5.2) ^b	(39) ^b (2.9) ^b (0.23) ^b (5.2) ^b	(5.5) ^b (23) ^b (47) ^b (0.27) ^b (1.6) ^b	(33) ^b 1.0 (0.93) ^b (0.55) ^b (3.2) ^b	2.2 2.3 1.0 (0.81) ^b (4.8) ^b

^aThese boundaries are selected such that the maximum annual (first-year) dose commitment to the critical organ will not exceed that recommended by the NCRP for annual occupational exposure.

The maximum acceptable dose commitment is not exceeded at any distance downwind. The numbers in parentheses are the maximum percentages of the maximum acceptable dose commitment, which occur 400 m downwind of the stack.

Table 8.18. Estimated Lifetime Dose Commitments to Critical Organs Resulting from Upper Limit Accidents at NFS, MFRP, BNFP, and Conceptual Plants for Processing LWR and FBR Fuels^{a,b}

				Conceptual	Conceptual LWR Plants of Capacity:	acity:	Conceptual	Conceptual FBR Plants of Capacity:	Japaci ty:
Type of Release	NFS	MFRP	BNFP	l Metric Ton/Day	6 Metric Tons/Day	36 Metric Tons/Day	1 Metric Ton/Day	6 Metric Tons/Day	36 Metric Tons/day
"Fresh" fission products Total number of fissions Thyroid dose commitment, rems Whole-body dose commitment, rems	10 ²⁰ ~2	10 ²⁰ 26 0.09	10 ¹⁸ 0.002°	2.7 × 10 ²⁰ 9.4 5.0	1.6 x 10 ²¹ 30 5.0	1.6 × 10 ²¹ 30 <u>5.0</u>	8.0 x 10 ²⁰ 0.65 5.0	1.6 x 10 ² 1 1.0 5.0	2.4 × 10 ²¹ 1.3 5.0
Noble gases (~Kr and ~~'Ke) Release, curies Whole-body dose commitment, rems	1 7	1 • 1	, 1	70,000 0,054	420,000 0.18	2,500,000	350,000 0.18	2,100,000 0.88	13,000,000 4.4
Halogens (*) and *(*)! Release, curies Thyroid dose commitment, rems	1.7	1.2	1.1	3.1 0.05	18 0.2	55 0.5	1100 4.6	6500 22	9700 27
Semivolatile fission products Release, curies 106 _{Ru} , curies Lung dose commitment, rems	1 1 1	1 1 1	1900 1500 ~0.0007°	760 410 2.7	4500 2500 8.9	4500 2500 8.9	3600 1300 5.0	7300 2600 7.9	11,000 3900 13
Nonvolatile fission products and transplutonics Release, curies 11μ (e, curies	1.1	W I	120 23	3.3	20 3.5	20 3.5	37.	74 4.7	111
Lung dose commitment, rems Bone dose commitment, rems		0.075	1.7 <0.0007° -	0.011 0.008 0.024 (0.005)	0.068 0.03 0.077 (0.017)	0.068 0.03 0.077 (0.017)	0.04 0.060 (0.024)	0.06 0.10 (0.04)	0.07
Plutonium Release, alpha curies Bone dose commitment, rems	59.0	133	0.11 <0.0007°	0.16 6.7 (0.26)	0.98 22 (0.8)	0.98 22 (0.8)	0.30 8.6 (0.3)	0.61 14 (0.5)	0.91
Distance to site boundary, km	1.5	9.0	~	ग ंग•	2.0	5.0	1.2	2.0	2.8

^aThe underlined numbers are those that fix the radial distance to the site boundary.

^bThe numbers in parentheses are the first-year dose commitment for those cases in which the first-year dose commitment is not equal to the lifetime dose commitment.

^CThe Allied Chemical Corporation reports the external exposure dose from beta and gamma radiation.

Fresh Fission Products. - Fresh fission products would be generated in a nuclear excursion. A nuclear excursion in a head-end vessel of maximum capacity, resulting in complete boildown of the solution, is assumed. After boildown and dehydration, the reaction would terminate in the assumed vessels because of the low effective density of the fissile material (~3 g per cm³ of uranium plus plutonium in calcined solids is assumed). The thermal power of the nonvolatile fission products (the fresh fission product heat is significant for the first 1 to 2 hr following the excursion) would then calcine the solids; these solids would probably subsequently melt through the vessel, flow onto the cell floor, and resolidify.

It is assumed that the initial rupture breaks the off-gas line and that all of the steam generated in the boildown phase (containing all of the noble gases, 30% of the iodine, 20% of the semivolatile fission products, and particulates of solution have the average concentration of nonvolatile fission products and plutonium) is discharged to the cell atmosphere and exhausted through the ventilation system. It is assumed that 99.5% of the semivolatile fission products are removed from the hot (air and saturated steam at ~100°C) ventilation stream by passage through metal mesh or silica gel absorbers. The ventilation systems of FBR plants are assumed to incorporate activated charcoal filters for removal of 9% of the iodine. The particulate release is calculated using the model presented in Sect. 8.3.2.

The doses delivered by a nuclear excursion are dominated by the whole-body dose that results from exposure to the radioactive cloud of fresh fission products (Fig. 8.17).

Release of Iodine Inventory. - It is assumed that a fire or explosion in a solid halogen absorber would completely release the contained equilibrium concentration of ¹³¹I and a two-year accumulation of ¹²⁹I. It is assumed that approximately 93% of the iodine collected by pretreatment in a wet scrubber is not dispersible. FBR plants are assumed to utilize charcoal filters that remove 9% of the remaining iodine.

The thyroid dose which results from the explosion of a silver reactor is obtained by properly prorating the ¹³¹I and ¹²⁹I doses at 0.4 km, as found in Tables 8.9 and 8.11, and applying the generalized dose curve (Fig. 8.15).

Release of Semivolatiles. - It is assumed that a total of 0.1% of the semivolatiles in the largest vessel is released by a mechanism other than a nuclear excursion (i.e., a tank boildown or an inadvertent addition of oxidants to a process vessel).

The upper limit accident in a waste tank for interim (2-year) storage of mixed fission products would release a smaller amount of ruthenium by comparison. In evaluating the waste tank accident, it is assumed that coolant is lost from the tank and that the tank leaks, discharging steam to the vault ventilation system and its condenser. The distillate, containing about 20% of the semivolatiles, is assumed to be returned to the tank, but an aerosol composed of particulates containing 20% of the concentration of semivolatiles in the waste is discharged through the filters. The release from this source is insignificant (semivolatiles content, <10⁻⁸ ton of fuel).

The doses resulting from the release of semivolatiles are controlled by the dose to the lung. They are obtained by application of the data in Tables 8.9 and 8.11, and the generalized curve (Fig. 8.15).

Release of Nonvolatiles. - The upper limit accident involving the release of nonvolatile fission products and transplutonic elements was determined to result from an explosion in the waste calciner containing fission products at a concentration of 10⁻¹⁴ ft³ per megawatt-day of burnup. The explosion in a waste evaporator would cause essentially the same release if the droplets evaporated in the ventilation system upstream of the filter. No "credible" accidents that would cause a larger release of nonvolatile fission products could be postulated in the interim solids storage pool or interim liquid waste tank.

The downwind doses resulting from the postulated releases were found to be negligible.

Release of Plutonium. - The maximum credible plutonium release was assumed to result from a fire of 0.5-hr duration (the fractional release is proportional to the time of aerosol generation) involving either resin or solvent loaded with plutonium. The particles escaping from the filter were assumed to be pure PuO₂ (a pessimistic assumption). The release of

plutonium from this source is about the same as that from a nuclear excursion involving complete boildown of the plutonium storage tank (containing up to 7.5 tons of plutonium).

The controlling bone dose from a plutonium fire is found in a manner similar to that used to calculate the dose resulting from the release of semivolatiles.

8.3.5 Maximum Theoretical Accident

A maximum theoretical accident has been evaluated for the purpose of illustrating the worst possible consequences that could result from poor design and/or implementation of good practice. Since waste storage tanks are known to have the largest inventory of physiologically hazardous materials, we have assumed that a hydrogen-air explosion occurs in the vapor space of an acid waste tank containing a 2-year accumulation of fission products.

Mechanical Consequences. - Illustrative (but not the worst possible) mechanical consequences of such an accident were made assuming that the tank contains fission products from 39,000,000 Mwd of fuel exposure (a 2-year accumulation from a 6-metric ton/day plant processing fuels irradiated to a burnup of 12,000 Mwd/ton), generating 56,000,000 Btu/hr in 390,000 gal of solution. The tank, 80% filled with solution, is assumed to: (1) be fabricated of 0.5-in.-thick stainless steel, (2) have a diameter of 65 ft and a height (with flat heads) of 20 ft, and (3) be housed in a 3-ft-thick concrete vault buried under 10 ft of earth. Following the loss of purge air to the tank, the concentration of hydrogen in the 13,000-ft3 vapor space would increase to 4 vol % (the minimum flammable concentration) after about 3 hr and to 30 vol % after about 24 hr. Assuming that the loss of purge air is undetected and that there is a source of ignition after 24 hr, the resultant explosion would liberate approximately 1,100,000 Btu of energy, generate a pressure of approximately 100 psig in the vapor space, elevate the concrete roof and earth cover by several feet, and (we assume) rupture the coolant and off-gas piping in such a manner that complete loss of cooling would ensue and the tank would be directly vented to the atmosphere.

Assuming that no remedial measures were taken following the loss of coolant and the breach of containment, the solution would heat to boiling in about 10 hr and evaporate to dryness after about 125 hr. The waste salts could calcine, melt through the floor of the tank, decompose the concrete, and flow into the earth beneath the tank after a total of approximately 160 hr following the explosion.

A comparable accident in an alkaline waste storage tank would have similar consequences, but would take place over a longer time period because of the greater dilution. Calculations made for a 1,200,000-gal alkaline waste tank containing waste generating 43,000,000 Btu/hr indicate that the waste would heat to boiling after approximately 23 hr, boil to dryness after approximately 273 hr, decompose after approximately 290 hr, and melt through the vault in approximately 330 hr.

The transient growth of a molten sphere in infinite media of dry sand and limestone was estimated, assuming that the fission products are mixed by convection in the molten zone and that the molten zone has the same density as the surrounding earth (so that no settling or flotation of the sphere would occur). Assumed properties of the dry sand and limestone are shown in Table 8.19. The results (Fig. 8.20) indicate that the radius of the molten sphere in dry sand would grow to a maximum of about 75 ft after about 1500 days. In limestone, the radius of the sphere would grow to a maximum of approximately 50 ft after 700 days. The molten zone would completely solidify after about 150 years.

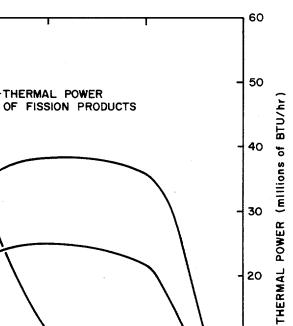
Release of Radioactive Materials to the Atmosphere. - The semivolatile fission products (Ru, Cs, and Te) would be released quantitatively to the atmosphere during the boildown-calcination phase of this maximum theoretical accident. In addition, about 0.1% of the mixed nonvolatile fission products would be released by entrainment in the steam; however, these can be neglected in an analysis of consequences because their effect is negligible by comparison. In the evaluation of downwind consequences, it can be assumed that thermal currents carry the semivolatile fission products to the top of an atmospheric inversion layer.

Table 8.19. Assumed Properties of Dry Sand and Limestone

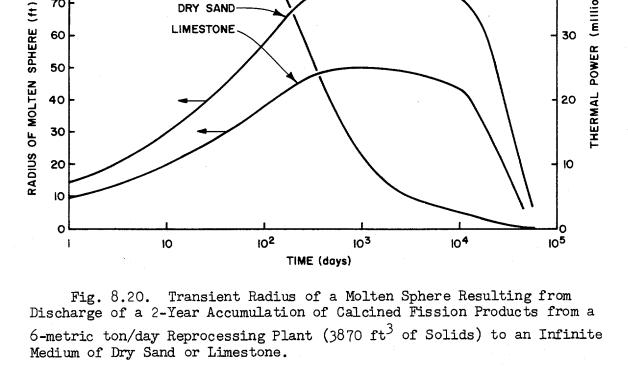
Property	Dry Sand	CaCO3	CaO ^a	Limestone
Density, o, lb/ft ³	6.49	162	90.8	
Heat capacity, c, Btu lb ⁻¹ (°F)-l	0.183 + 0.0000µ46T	0,203 + 0,0002T	0.177 + 0.00011T	
Thermal conductivity, k, Btu hr ⁻¹ ft ⁻¹ (°F)-1	0.18 + 0.00012T	1.3 + 0.00012T	0.18 + 0.00012T	
Transition temperature, T , $^{\circ}F$	3133 (melt)	1 μ 00 (loses κ 02)	4660 (melt)	4660 (melt)
Heat of transition, &, Btu/lb	50.1	0.0	385	
Average thermal diffusivity, α , ft ² /hr	ሳ.0.01			0,011
80, Btu/ft3	4750			35,000
$(T_m - T_a) c/\ell$	15.5 ^b			6.37 ^b

Ande by calcination of $CaCO_3$.

banbient earth temperature, $extsf{T}_{\mathbf{a}}$, is 77°F.



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80

70

60

50

40

DRY SAND LIMESTONE The release to the atmosphere from the maximum theoretical accident in an alkaline waste tank would be comparable to that from the acid waste tank except that it would take place over a longer time period. The cerium would probably be released quantitatively during the boildown phase. The ruthenium would be released during the calcination phase, although it is probable that a portion would be deposited on cool (<800°C) surfaces as it diffuses to the postulated break in the off-gas line.

The consequences of a waste tank boildown may be found by proper prorating of the 0.4-km doses found in Table 8.13 and by use of the generalized dose curve (Fig. 8.18). The doses thus obtained (>10⁵ rems at 0.4 km downwind) have little meaning other than to show why such maximum theoretical accidents must be rendered incredible through the use of appropriate engineered safety features.

Another related type of accident in an acid waste tank, also considered only of a theoretical nature since it depends on a very improbable combination of circumstances, involves simultaneous failure of the coolant for the coils of the tank and the off-gas condenser. In the event of such an accident, the contents of the tank would boil down on essentially the same time scale as that discussed previously. Because of the low heat capacity of the air and typical ventilation ducts, a mixture of air and saturated steam at approximately 100°C could pass through the off-gas and ventilation filters and be exhausted to the stack. Certain of the semivolatile fission products (in particular, ruthenium tetroxide, which has a boiling point of ~80°C) may be carried by this stream. It is assumed (as in the case of the nuclear incident discussed previously) that 20% of the ruthenium is volatilized during the boildown phase and that, of this, 99.5% is removed by deposition on metal or on the filter. remainder of the semivolatile fission products might be evolved in the calcination phase, but the off-gas line is assumed to cool following cessation of steam flow, permitting essentially complete (by comparison) removal of the semivolatile fission products by deposition and filtration.

Mitigation of Accidents. - The authors stress that such accidents as the one denoted as "maximum theoretical" may be converted to the tolerable category, in terms of consequences, by proper forethought and design. For

example, the effects of the postulated hydrogen-air explosion can be mitigated by one of the following (and possibly by others, limited only by the ingenuity of the designers):

- (1) Increase the reliability of preventive measures for control of the purge air flow and the hydrogen concentration.
- (2) Enclose the waste tanks within a building that is ventilated through a condenser and filter.
- (3) Design the tank and/or the vault to withstand a pressure of about 100 psig without rupture.
- (4) Decouple the tank from the vault. Use a pressure suppression and/or pressure relief system in the tank. Vent the vault to a containment system with large capacitance or to a pool of water for steam suppression.
- (5) Use titanium tanks and self-boiling wastes to ensure effective purging of the hydrogen by steam.

8.3.6 Consequences of the Leakage of High-Level Wastes to the Ground

Radioactive waste solutions that are released by tank failure might be routed through the geologic formation lying between the tank site and the nearest surface drainageways. Since analyses must be made using specific site conditions, a hypothetical tank site at Oak Ridge was chosen for didactic purposes. This site was considered to be located in Conasauga shale on a promontory, with intermittent surface streams passing to the east, south, and west of the tank site. The shale formation is quite impermeable, and the movement of water is restricted so that it flows only along bedding planes.

Samples of the Conasauga shale were obtained below the highly weathered zone in a direct path toward the surface streams. These samples were acidified for the removal of calcite, and the exchange capacities were determined by the calcium titration method of Jackson. A mean value of 11 ± 1 meq/100 g was obtained. Overnight refluxing in $7 \, \text{M}$ HNO3 at $85 \, ^{\circ}\text{C}$ showed a hydrogen ion consumption of $260 \, \text{meq/100}$ g, which would be sufficient to neutralize

the entire contents of an acid waste tank within a distance of 30 ft from the tank. In the case of acid waste, it was assumed that neutralization of the acid by calcite in the formation would result in a calcium salt system. In this system, strontium was assumed to compete with calcium without selectivity of either ion, although strontium might be slightly more selectively sorbed than calcium. For the sorption of strontium from neutralized wastes, and for the sorption of cesium and ruthenium, information on the sorption properties of Conasauga shale were obtained from previous laboratory studies. 78-82

The quality of the groundwater was assumed to be similar to that of Clinch River water, which has a total cation (calcium and magnesium) concentration of about 0.002 meq/ml. Seepage rates were assumed to be characteristic of the area surrounding Waste Pit 2, where the average seepage rate from 1953 to 1958 was 3900 gal/day through an average sidewall area of 9000 ft² (ref. 84). This corresponds to a mean superficial velocity of 0.064 ft/day. A mean groundwater velocity of 0.67 ft/day was used, which implies approximately 10% efficiency of contact between the shale and solution. If the initial seepage rate were maintained, the daily seepage rate from the acid waste tank (filled to a height of 35 ft with 10⁶ gal of waste) would be 2275 gal. The seepage from the neutralized waste tank (filled to a height of 36 ft with 1.25 x 10⁶ gal of waste) would be 23h0 gal.

Dispersion properties of solution in the formation (Fig. 8.21) were estimated from the results of a chloride tracer test conducted at the site. 84 These data indicate an effective plate height of 46.5, according to the notation of Glueckauf. 85

Calculation of Radionuclide Movement. - In addition to the assumptions outlined above, it was further assumed that the waste would move longitudinally through a zone 75 ft wide, with a height equal to the original liquid level in the waste tank, to surface water at a distance of 200 ft. No allowance was made for lateral dispersion, but the spread of the solute was assumed to occur according to Glueckauf's model for the elution of a band of solute through a linear ion exchange column. The porosity of the

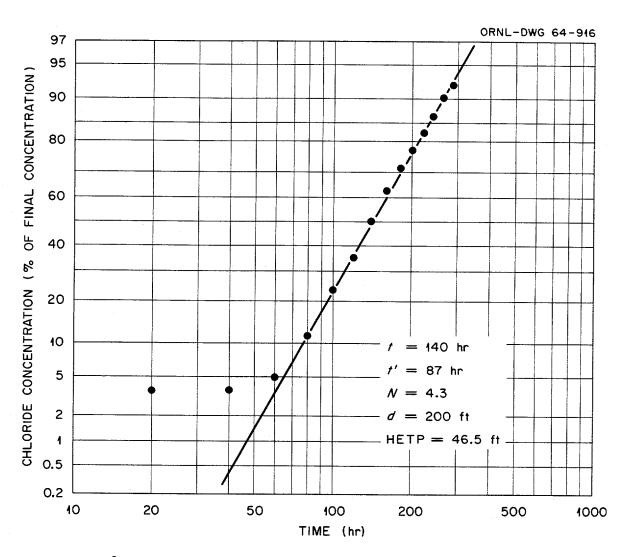


Fig. 8.21. Dispersion Properties of Chloride in Conasauga Shale at a Four-Acre Tank Site.

shale effectively contacted by solution was assumed to be 25%, with a grain density of 2.64 g/ml.

If a leak were to develop in a waste tank, the amount of solution lost to the formation would be limited by the ability of the formation to accept the solution. During percolation of the waste solution, the groundwater concentration in the zone of migration would be increased, returning to normal when the waste solution was again displaced by the local groundwater. Movement and dispersion of the specific radionuclides were estimated by using Glueckauf's model in order to describe the dispersion of the unsorbed anions and correcting for retention of the radionuclides by the formation, as discussed by Inoue and Kaufman. However, due to the variable concentration of electrolyte in the groundwater, the retention factor was not constant with time. In addition, radioactive decay was considered.

The results of calculations for the movement of 90 Sr from an acid tank are shown in Fig. 8.22. The initial peak in 90 Sr activity at the surface drainageway occurs at about 1 year and is due to the relatively slight sorption of strontium by the shale in the presence of high concentrations of electrolyte. With time, these high concentrations of salt are diluted and replaced by fresh groundwater, and a second concentration peak occurs after about 150 years. The relative magnitude of these two peaks depends on the total quantity of electrolyte released to the formation. If, after a leak occurs, the waste solution is pumped from the ground, the initial rapid movement will not be observed due to the removal of the excess electrolyte. Furthermore, in the case of 90 Sr in an acid waste system, an appreciable fraction of the total radioactivity could be removed (Table 8.20).

For neutralized waste, the precipitation of strontium, in addition to the increased probability for ion exchange, prevents 90 Sr from attaining any significant concentration at the surface drainageways. The high affinity of the Conasauga shale for cesium deters movement of 137 Cs so that radioactive decay occurs before significant concentrations would be observed in either acid or neutralized waste systems. The relatively rapid

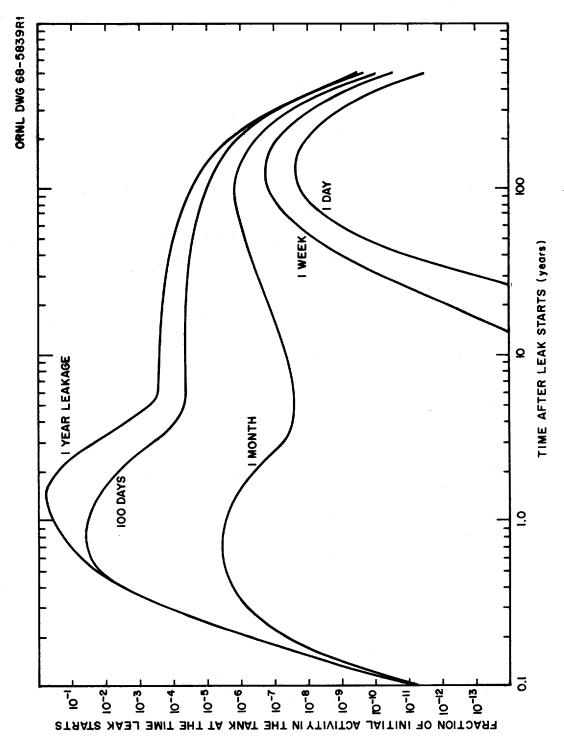


Fig. 8.22. Strontium-90 Activity in the Groundwater at a Point 200 ft from a Leaking Tank of Acid Waste.

Table 8.20. Recovery of Radionuclides from the Soil After a Leak Has Developed in a Waste Tank

	Percent	cage Recoverable
Isotope	Acid Waste	Neutralized Waste
90 _{Sr}	88	<1
137 _{Cs} 106 _{Ru}	2	<1
106 _{Ru}	18	20

decay of 106Ru (half-life, 1 year) would prevent it from attaining significantly high levels at surface seeps unless a very extensive leak were to occur.

Several factors must be incorporated into the mathematical model in order to arrive at predicted concentrations. Each parameter used is subject to variation. Figure 8.22 was estimated using the following guesses for these parameters:

Mass of soil contacted per milliliter of pore solution	8 g
Stable composition of acid waste	5.7 M
Stable composition of groundwater	
Distance of travel	
Groundwater velocity	0.67 ft/day
Theoretical plate height	
Strontium distribution factor	

In addition, cases were considered in which each of these variables was allowed to vary by ±10% of its average value. The maximum and minimum concentration curves for a 100-day leak are shown in Fig. 8.23. It is seen that, although the shape of the curve (as plotted on a log-log scale)

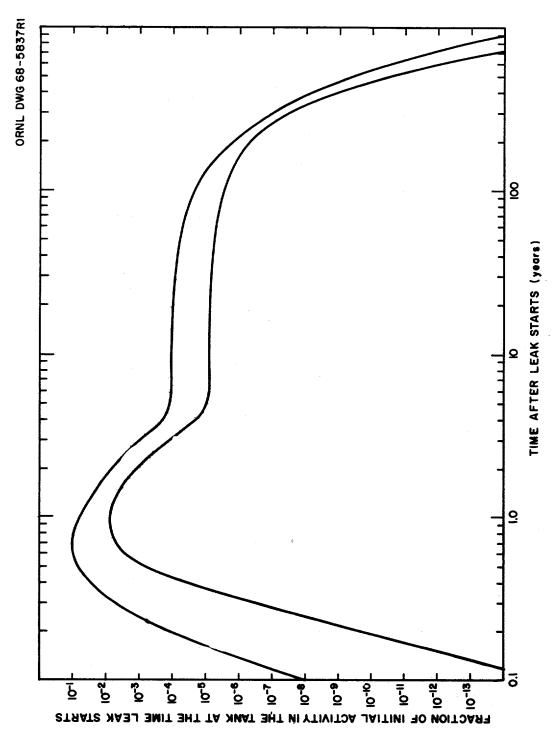


Fig. 8.23. Strontium-90 Activity in Groundwater Resulting from 100 Days of Leakage from an Acid Waste Tank.

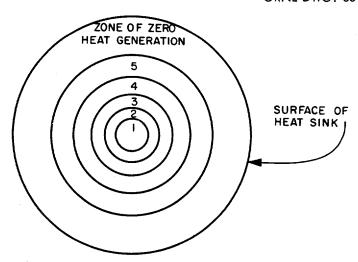
is not significantly affected, estimates of the activity levels at any time may be in error by one or two orders of magnitude.

The distribution factor and the mass of soil contacted per milliliter of pore solution will determine the relative velocity of the radionuclide in comparison to the percolating groundwater. The larger these two factors, the slower the relative velocity. Since the distribution factor is inversely proportional to the concentration of the percolating solution, the movement of the radionuclide is most rapid initially and is then reduced as the groundwater concentration returns to normal. The level plateau on the curves between the two peaks reflects the ratio of the normal groundwater concentration to the concentration of the waste solution.

The time of travel is dependent on the groundwater velocity and the distance traveled. In our local situation, flow is nearly unidirectional However, because this would not likely be the case in other situations, additional consideration would need to be given to the geometry of the flow patterns. In our situation, it was found that the dispersion of the chloride tracer could be described by a chromatographic breakthrough curve with a theoretical plate height of 46.5 ft. This implies that about 1% of the groundwater will traverse 200 ft in one-third the time of the average movement. In other situations, the average groundwater velocity may be quite different from the values we used; thus it may be impossible to fit travel times to simple dispersion or chromatographic breakthrough equations. A greater degree of dispersion hastens the appearance of radioactivity at a given point, but the peak concentrations are diminished unless adequate time has elapsed to permit radioactive decay.

Thermal Effects. - The distribution of radionuclides in the soil is important because of the thermal problems that are likely to result from high concentrations of radioactive material in a medium that has poor heat-conducting properties. Spherical shell geometry and thermal equilibrium were assumed as a first approximation to estimate the magnitude of the thermal problem in contaminated Conasauga shale. Figure 8.24 shows the simplified model and the heat-generating capacity of the contaminated shale according to zones. A solution in the spherical shell geometry has been described by Etherington. In the present study, the tank has been

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	LIQUID F	RETAINED	LIQUID NO	T RETAINED
ZONE	Btu ft ⁻³ hr ⁻¹	ft³/gal	Btu ft ⁻³ hr ⁻¹	ft ³ /gal
1	200	0.15	61	0.15
2	280	0.05	220	0.06
3	250	0.09	140	0.25
4	33	0.18	4.7	2.80
5	0.45	0.01	0.05	0.73

Fig. 8.24. Configuration and Characteristics of Contaminated Soil Zones.

ignored as a heat sink, and a heat sink was assumed to be located at the edge of a variable-sized non-heat-generating region. To define the limits of temperature rise, two cases were considered. The first assumes that liquid waste is retained in the shale pore volume, and the second assumes that liquid is not retained in this way. The source strength is listed for each zone, and the volume occupied by each zone is normalized to 1 gal of waste. For example, if a leak of 1000 gal occurs and liquid waste is retained by the shale pores, then the volume of zone 1 is 150 ft³ and the heat-generating rate in this zone is 30,000 Btu/hr.

When acid waste leaks from a tank, it will be neutralized by the buffering action of the shale (260 meq of H⁺ per 100 g of shale). Zone 1 depicts the acid zone; the remaining zones are neutralized. Soil loadings were estimated from studies conducted at ambient temperatures and from judicious application of these results to the analysis. Values of thermal conductivity of unweathered Conasauga shale range from 1 to 2 Btu min⁻¹ ft⁻¹ (°F)⁻¹ when measured at 30 to 33°C. Similar data do not exist for elevated temperatures. Therefore, it was assumed that the thermal conductivity of shale increases gradually with increasing temperature and follows the pattern observed in metal systems. In all likelihood, the thermal conductivity of shale will decrease as the temperature increases, especially when water is lost from the shale. Thus, the temperature rise estimated in this analysis is likely to be lower than that which may actually occur.

Figure 8.25 shows the steady-state temperature attained in the center of the spherical system as a function of leak volume. Maximum temperatures increase with the volume of the leak, the retention of liquid in pore spaces, and the absence of a heat sink. Temperatures range from 870 to 1250°F for a leak of 100 gal.

Transient temperatures are being investigated as a function of time and space in a system that includes a variable-sized cylindrical geometry (representing the contaminated zone) and variably spaced heat sinks located at the tank, the groundwater table, and the ground surface.

A digital computer program, TOSS, contains many of the requisites for solving the transient problem for a multivariate system. 90 This program,

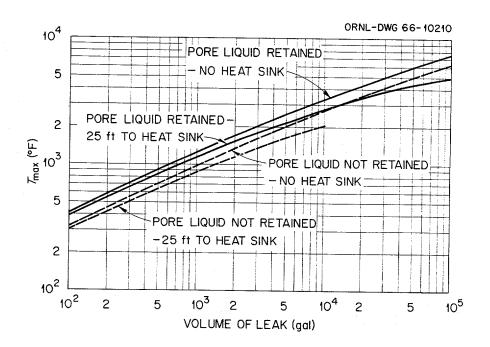


Fig. 8.25. Steady-State Temperatures Attained in the Center of the Spherical System as a Function of Leak Volume.

which has been modified to suit the needs of the present study, has the capability of calculating the temperature distribution for a three-dimensional, multiregional problem having internal heat generation. A number of cases are being analyzed. Studies are also under way to evaluate the diffusivity, or specific heat, and the thermal conductivity of Conasauga shale as functions of temperature.

Conclusions. - In a formation similar to Conasauga shale, the slow rate of percolation of the solution, combined with rather high sorptive properties of the formation (except for 90 Sr in an acid waste system), would tend to prevent the rapid release of large quantities of radionuclides directly into surface waterways. However, this delay would result in the buildup of activity in the formation to levels that would probably present a serious thermal problem. The delay time afforded by the formation could be used for remedial measures (e.g., for pumping groundwater from the formation to recover the unsorbed radionuclides and for preventing further transport of the fission products).

The absolute values for radionuclide movement that have been calculated and presented in this discussion should not be considered to be precise since the estimates were based on a rather limited description of the site. However, the procedure for making these estimations could be applied for any proposed site. Actual tests of seepage and dispersion at a proposed site employing the layout of the tank system would provide a more adequate basis for such calculations.

8.4 Requirements for Treatment of Routine Effluents

The preceding sections have shown that the assumed routine releases of radionuclides from fuel reprocessing plants require greater site boundary distances than those required by the assumed upper limit accidents. The large site boundary distances that were estimated to be required by the routine release from plants of high capacity provide incentive for partial removal of the noble gases and a larger percentage of the iodine than that assumed in Sect. 8.2.1.

Table 8.21 shows approximate site boundaries that would result if

Table 8.21. Required Factors for Removal of Noble Gases and Iodine Such That the Site Radius Is Controlled by the Upper Limit Accident

			Reprocessing 1	Reprocessing Plant Capacity (metric tons/day)	ric tons/day)	
		LWR Fuel	-1		FBR Fuel	
	Ľ	9	36	Т	9	36
Site radius, km	0.144ª	2.0	2.0	1.2	2.0	2.8
Site area, km ²	19.0	12	12	4.5	12	25
Noble gas DF ^b	0.58°	2.6	16	4.4	20	92
Iodine DF ^b	150	089	700	5.2 × 10 ⁶	2.4×10^7	1.1 x 10 ⁸

 $^{
m a}{
m At}$ the ORNL site, the required DF's are unchanged for site boundaries less than 1.2 km to the northeast since the average annual downwind concentration "peaks" at this distance.

that required to attain, at the site boundary, average annual concentrations of 1 $ext{x}$ 10-7, 0.77 $ext{x}$ 10-13, and 1.4 x 10^{-13} curies/m³ of noble gases, iodine from LWR fuel, and iodine from FBR fuel respectively. ^bDF is defined as the ratio of the average annual release rate obtained without treatment devices to

^CNo removal of noble gas is required. The average annual noble gas concentration at the site boundary is $5.8 \times 10^{-8} \text{ curies/m}^3$. the noble gases and iodine were removed from the normal effluent to such an extent that the maximum site radius is determined by the upper limit accident. On this basis, equipment for removing 50 to 99% of the noble gases appears to be necessary for plants with capacities of more than a few tons per day. More efficient iodine removal than that demonstrated in present technology will be required for LWR plants with capacities greater than about 6 to 10 tons/day, and decontamination factors (DF's) as high as 10^8 will be required for FBR plants if the spent FBR fuel is to be processed after decay times of only 30 days.

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